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PERFORMANCE OF ENGINEERED BARRIERS FOR LOW-LEVEL WASTE

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ABSTRACT

Geotechnical Resources Ltd., in association with Komex Consultants Ltd., was retained to collect, synthesize and evaluate the available information on the long term performance of engineered barriers for low-level radioactive wastes disposed in Canada. Literature was researched from Canadian, United States and European sources. A variety of barrier materials were assessed in the study and included natural clays, concrete and cement, metals, bentonite-sand admixes, bitumen and bituminous admixes, soil cement and polymeric membranes. The generalized geological and geotechical conditions encountered within the soil and rock host media currently under consideration for disposal sites in southern Ontario were also summarized.

Both internal barriers, or buffers, to immobilize the waste material and reduce radionuclide mobility, as well as external barriers to limit the migration of contaminants were examined. Microbial activities within the waste forms were analyzed, including cellulose degradation, methanogenesis and bicarbonate and organic reactions. Microbial interactions with the various engineered barrier materials under consideration were also assessed.

Finally, the anticipated long term performances of the respective barrier materials under consideration were evaluated, along with the general suitability of the geological host media being proposed for disposal sites.

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RÉSUMÉ

Dans le cadre de ce projet, Geotechnical Resources Ltd., en association avec Komex Consultants Ltd., a assemblé, synthétisé et évalué l'information disponible sur la performance à long terme des barrières artificielles pour contenir les déchets de faible activité radioactive qui sont enfouis au Canada. Un examen de la littérature technique sur le sujet a été effectué. Plusieurs sources d'information en provenance d'Europe, des États-Unis et du Canada ont été consultées. Dans cette étude, plusieurs matériaux utilisés pour la fabrication des barrières ont été évalués. Ces matériaux incluent: les argiles naturelles, les bétons et les ciments, les métaux, les mélanges de sable et bentonite, les mélanges à base de bitume, le sol-ciment et finalement les membranes polymériques. Les conditions géotechniques et géologiques rencontrées dans les sols et la roche des milieux d'enfouissement présentement considérés pour l'enfouissement des déchets de faible activité radioactive dans le sud de l'Ontario ont aussi été synthétisées.

Les barrières internes ou zones tampons qui immobilisent les déchets radioactifs et réduisent la mobilité des radinucléides ainsi que les barrières externes qui limitent la migration des contaminants ont été examinées. Les activités microbiologiques prenant place à l'intérieur de la masse (forme) des déchets radioactifs ont été analysées. Ces activités incluent: la dégradation cellulosique, la production de méthane, les réactions organiques et les réactions bicarboniques. Les interactions microbiologiques avec les différents matériaux de construction des barrières considérées dans cette étude ont aussi été évaluées.

Finalement, les performances anticipées à long terme des matériaux de construction des barrières ont été évaluées, de même que la pertinence du milieu géologique proposé pour les sites d'enfouissement.

NOTE:

La Commission de Contrôle de l'énergie atomique n'est pas responsable de l'exactitude des opinions ou des énoncés exprimés dans ce rapport. La Commission et les auteurs n'assument aucune responsabilité pour les pertes et les dommages qui peuvent être encourus suite à l'utilisation de l'information contenue dans cette publication.

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1. INTRODUCTION AND SCOPE OF STUDY

1.1 Scope of Study

Geotechnical Resources Ltd., in association with Komex Consultants Ltd., both of Calgary, Alberta, were retained by the Department of Supply and Services on behalf of the Atomic Energy Control Board to gather and evaluate available information relevant to the long term performance of engineered barriers for low-level wastes (LLW). The project consisted of the following tasks:

a) A brief summary of the engineered barrier concepts in practice, or under consideration, for shallow low-level waste disposal in Europe, the United States and Canada.

b) Compilation, review and synthesis of available information pertaining to the long term performance of engineered barriers being considered in Canada for low-level waste.

c) Summary of the geological and geotechnical conditions at defined potential shallow low-level waste disposal sites in southern Ontario and relationship to barrier concepts under consideration.

d) Assessment of the probable durability of barriers, factors affecting their integrity and overall anticipated long-term performance.

For the purpose of this study, engineered barriers are considered to include both internal and external engineered barriers. External barriers are comprised of both structures to limit the migration of contaminated liquids away from the waste unit, as well as covers to minimize the infiltration of water into the unit. Internal barriers are

used to solidify and immobilize the waste material. Waste form is the radioactive waste material including the package material and inherent microbial population. In addressing the performance of barriers, susceptibility to physical breakdowns, chemical and biological attack and biotic intrusion was considered.

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a) project administration - Mr. Alain Pare, Geotechnical Resources Ltd.

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c) hydrogeological aspects - Dr. T.L. Dabrowski, Komex Consultants Ltd.

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Prior to finalizing the report, the findings from the study were presented at a debriefing meeting held in AECB headquarters in Ottawa on April 22, 1987.

2. LOW-LEVEL WASTE - A CANADIAN PERSPECTIVE

At the present time, there appears to be no clear definition for low-level waste; most definitions being by exclusion. Coleman and Dodge[1], reflecting the USA point-of-view, defined low-level waste as "radioactive material that is not high-level radioactive waste, transuranic waste, spent nuclear fuel or by-product material". A similar definition for low-level waste has been cited by Cameron et al[2] of the Low-Level Radioactive Waste Management Office (LLRWMO) who defined low-level waste as "all radioactive wastes except those produced by uranium mining and milling, and the irradiated nuclear fuel from nuclear reactors". Both of these definitions imply that low-level waste includes both low and intermediate radioactive waste material.

Feraday[3] indicated that the activity of low-level waste is less than 0.1 Ci/m^3 (/). In fact, due to incineration, compaction and volume reduction processes, the activity within the waste repository will likely be in the order of 1 Ci/m^3. For the purpose of this study, the LLRWMO definition for low-level waste has been adopted to reflect the Canadian perspective. This definition implies that a small percentage of the waste (ion exchange resins and columns) may have an activity level of 200 Ci/m^3. Nevertheless, the majority of the waste will be in the order of (less than) 1 Ci/m^3, as indicated in Feraday[3].

Cameron et al[2] have catagorized low-level waste producers into four main groups. Table 1 presents a summary of the low-level waste characteristics within each of these groups. Information listed in Table 1 includes source of waste, typical wastes, annual production and radionuclide type and activity level, where available. Radionuclide type and activity level were extracted from Buchnea et al[4]. It should be noted that the volumes listed in Table 1 do not include historic waste, such

as the 1625 m^3 generated by Eldorado Resources Limited (ERL). Also to be noted is that the waste produced by the radio-isotope users (Group 1) is presently stored at Chalk River Nuclear Laboratories (CRNL).

It is anticipated that between 8,000 and 10,000 m^3 of low-level radioactive waste will be produced annually in Canada[2]. The province of Ontario produces over ninety percent of this radioactive waste with Ontario Hydro (35 percent), CRNL (28 percent) and ERL (14 percent) being the main generators. The vast majority of this radioactive waste is solid waste material. Liquid wastes include tritiated water and oil[5] and CANDU reactor decontamination waste such as TURCO 4512, potassium-persulphate and peroxide bicarbonate[6].

In terms of engineered barrier performance, the most important radionuclides are Cesium-137, Strontium-90, Cobalt-60, Tritium and Carbon-14, which have half lives of 30.2, 28.1, 5.3, 12.3 and 5730 years, respectively. C-14, is associated with ion exchange columns. Other radionuclides will be part of the waste form. These radionuclides have short half-life values (less than 1 year) and therefore may not require engineered barriers because storage for 10 half lives reduces radiation level 1,000 fold.

At present, little or no information is available on the chemical composition of the radionuclides. Personal communication with CRNL personnel suggests that the chemical concentrations of radionuclides within the waste forms are extremely low and therefore are not expected to have a significant effect on engineered barrier performance.

TABLE 1: SUMMARY OF LOW-LEVEL RADIOACTIVE WASTE

PRODUCER GROUP	SOURCES	TYPICAL WASTES	VOLUMES m^3/annum	RADIONUCLIDE TYPE (MAIN) AND ACTIVITY RANGE TBq/a
1	. Radio-isotope Users (research institutions and hospitals).	. Sealed sources of undisturbed equipment such as gauges, radiography cameras, static electricity eliminators. . Contaminated materials such as animal carcasses, scintillation vials, liquids, filters, syringes, wipes and gloves.	322.5	140 Co-60 15 Cs-137 7 Eu-154 11 Pm-147 other mixed isotopes
	. Nuclear Fuel Fabricators		370.0	
	. Eldorado Resources Limited (ERL)	. Uranium contaminated materials and residues (large historic waste volumes).	1625.0	1.5 Uranium
2	. Nuclear Utilities, e.g., Ontario Hydro, Hydro-Quebec and New Brunswick Electric Power Commission	. Reactor purification system waste such as filter vessels, ion exchange columns resins. . Reactor maintenance waste such as paper, cloth, glass, plastic, sheet metal materials, piping and equipment components.	3912.5	150 H-3 1.6 Cs-137 .4 Sr-90 .4 Cs-137
3	. Incidental Wastes	. Residues from abrasive manufacturing or speciality metal alloy production (phosphogypsum tailings, filters and tank liners are <u>not</u> included).	1427.5	.003 Ra-226 .003 Th-230 .05 Th-232 .05 U
4	. Atomic Energy Canada Ltd. (AECL), e.g., CRNL and WNRE	. Contaminated material from laboratory and plant operation, reactor maintenance and purification wastes and wastes from isotope.	1530.0	3.7 H-3 2.6 Co-10 0.5 Sr-90 0.5 Cs-137

3. OVERVIEW OF PRESENT DISPOSAL PRACTICES AND ENGINEERED BARRIER CONCEPTS FOR LOW-LEVEL WASTE

3.1 U.S.A and Europe

This section is included to provide an international context in which Canadian low-level waste disposal practices can be viewed. Data presented in this section were extracted from the proceedings of the "International Seminar on Radioactive Waste Products - Suitability for Final Disposal", held in Julich, West Germany[7].

All solid and liquid low-level waste to be disposed will follow the "concentrate and contain" philosophy. The "multiple barrier" principle for repository design is essentially universal. However, there is a significant variety in the methods and levels of development in low-level waste disposal among the countries considered.

A summary of the low-level waste disposal practices and plans for eight countries active in the nuclear industry is presented in Table 2. Several key aspects of the disposal system are considered, with emphasis on the geologic (host media) and engineered barriers.

The majority of radioactive waste produced in the U.S.A. and Europe comes from "light water" nuclear power reactors. These reactors produce significantly different waste than the Canadian "heavy-water" (CANDU) reactors. Hence, comparisons between Canadian and other countries' practices for disposal of low-level waste should recognize this difference.

Of all the countries considered, only the U.S.A., Sweden and Italy have chosen the shallow soil burial approach. Belgium plans to develop a repository in a deep clay stratum. All other countries, except the

Netherlands, which has not finalized its plans, are using or will use bedrock host media for deep underground repositories.

Several other key points, evident from the data contained in Table 2, are discussed as follows.

All countries use at least one type of waste processing method. These operations focus on volume reduction and immobilization.

Volume reduction (compaction, incineration) significantly improve the efficiency of LLW disposal operations by greatly reducing the quantity of material that must be stored, while keeping radiation levels within a manageable range.

Immobilization is typically done with concrete, cement or bitumen. These materials form a matrix with the processed LLW and provide an internal barrier to radionuclide migration.

LLW package production is a manufacturing process in every sense. For safety reasons the various processes used are carefully controlled and monitored. This is indicated qualitatively in Table 2 under the headings of "waste acceptance criteria" and "waste package quality assurance system".

In all cases considered in Table 2, LLW is loaded into containers before placement in the repository. These containers or containor systems form an external barrier to the waste form. Steel containers (sealed), concrete containers, external concrete layers (overpacks) or combinations of these are typically used.

External to the containerized LLW, more variation is seen in repository and barrier types. In shallow burial cases (U.S.A., Sweden in part, Italy) repository backfill (buffer) materials and structural barriers

are more important than in deep respositories. In the latter cases, such as the bedrock repositories of Sweden, Finland, West Germany and Switzerland and Belgium's proposed deep clay stratum site, the host media are considered to play a larger role in overall repository performance and long-term safety. In the Swiss case, especially all long-term containment is attributed to the bedrock host media. This approach necessitates that considerable geological and engineering effort be focused on the complex subject of radioactive waste containment in geologic host media.

3.2 Canada

Over 99 percent of the low-level waste produced in Canada is being stored at facilities operated by Atomic Energy of Canada Limited (AECL) at its Chalk River Nuclear Laboratories (CRNL) site, or by Ontario Hydro. Other low-level wastes produced by hospitals, industry and universities (Toronto and McMaster) are transferred to the Chalk River facility. Some low-level waste is produced during uranium refining by Eldorado Resources Ltd.; current disposal plan is presently being reviewed. Hence, the following sections address the disposal practices and engineered barrier concepts being addressed by AECL and Ontario Hydro.

3.3 Plans by Atomic Energy of Canada Limited at the Chalk River Site

The favoured LLW disposal concept under consideration by Atomic Energy of Canada Limited (AECL) is emplacement of the waste in a shallow land burial (SLB) repository, which would ideally consist of trenches bounded by concrete walls, with a concrete cap and a permeable, but adsorptive floor. The hazardous lifetime for waste disposed into a SLB is designed for less than 500 years. An alternative concept for less hazardous waste, with a lifetime of up to 150 years is disposal into improved sand trenches (IST)[8].

AECL plans to condition waste by immobilizing incinerator ash, salt and ion exchange resins with bitumen. Attention is also being given to containing the waste in appropriate packaging, such as concrete overpacks and metal containers, as an external barrier to radionuclide migration. Concerns, however, have been expressed about the long term durability and corrosion resistance of such containers[9] and therefore their usage would require investigation.

Other materials under consideration, and which have been tested, are bitumen, cement and fine silica. Buckley[10] reported that diffusion coefficients are reduced by several orders of magnitude for all of these materials, including the silica which caused a reduction in the diffusion coefficient by a factor of 2000. In the latter case, although the silica has no capacity to adsorb radionuclides, it is effective in reducing the driving force for diffusion by decreasing the surface area of the waste in contact with water and by increasing the concentration of the bulk phase.

The barrier types under consideration by AECL are summarized in Table 3. Backfill and buffer materials which have been proposed include the following:

a) local sand
b) sand-clay mix (10% clay and 90% local sand)
c) illite, sodium bentonite, kaolin and attapulgite clay

Cover materials under consideration include reinforced concrete, plastic membranes and different types of soil. Incorporation of at least one layer of "heavy plastic sheeting", such as hypalon, is suggested[3,9]. Soil cover would consist of layers of sand, gravel, cobbles and boulders. The use of low permeability clay, in conjunction with pervious drainage layers to provide a "wick" effect is under consideration[3]. A central layer of asphaltic concrete has also been proposed.

Where concrete walls and covers are used, it is proposed to apply sealants to the construction joints, along with epoxy or bituminous coatings. Also, application of a polymeric sheet on top of concrete is under consideration.

In the trench bottom, a layer of drainage gravel is proposed to be placed directly over the native sand. On top of this layer, a 1 m thick layer of bentonite and sand is suggested to serve as an adsorptive layer (buffer)[3].

To reduce the effects of subsidence, it is proposed to construct the engineered cover 8-10 years after the end of filling. Concrete would be poured over a clay dome placed above the waste. A layer of fine-grained soil would be placed over a pervious layer, which would serve as a "wick". An impervious barrier, such as hypalon, would be placed over this layer which in turn would be overlain by layers of clay and topsoil. Under this concept, the concrete cover would be below the depth of seasonal frost penetration. The floor of the repository is permeable to maintain the waste in as dry a condition as possible, on the assumption that the water table will remain below the repository floor.

3.4 Plans by Ontario Hydro

At present, Ontario Hydro is storing its LLW and ILW in concrete cells, both in and above the ground. Practices over the past 15 years are described in the paper by Carter and Rao[11]. Although Ontario Hydro plans to store its wastes for the forseeable future, concepts for ultimate disposal have been developed[11,12,13]. The following description of engineered barrier concepts under consideration is based on these references. The planned depth of disposal ranges from shallow (understood to be within the upper 8 - 10 m) to intermediate (which is understood to be up to 100 m). Both soil and rock materials are under

consideration as the host media. Barrier concepts for disposal in both types of media are discussed in the following.

A summary of barrier types under consideration by Ontario Hydro is presented in Table 4.

3.4.1 Disposal in Rock Media

The LLW will be emplaced directly into the rock; the waste form probably being left in containers of metal or concrete which are transported to the respository. Depending on the level of radioactivity, consideration will be given to backfilling voids between LLW containers with clay till or soil-rock-bentonite mixtures. For example, the lowest three layers of containers placed in the cavern could be backfilled. In addition, the entrance to a tunnel or cavern unit would be plugged with low-strength flyash cement, otherwise known as K-crete. For ILW, the use of dry sand - bentonite is also proposed for filling voids.

3.4.2 Disposal in Soil Media

LLW may be placed either into trenches or tunnels. Preference is given to placement in the unweathered zone, below the surface zone of fissured soil. It is also intended to provide for a sufficient buffer between the bottom of the disposal unit and the bedrock surface, or potential aquifer, to retard nuclide migration. Adequate cover on top of the unit is also desirable. All of these criteria may not be easy to satisfy at a particular disposal site.

One concept is to enclose the waste in a concrete structural unit, with a slab cover supported on structural walls. This would reduce subsequent subsidence effects.

No external barriers are proposed, other than what is provided by the host media and the engineered cover. However, low permeability backfill will be used to restrict the rate of nuclide migration. Low strength flyash concrete backfill (K-crete) would be poured after each layer of LLW containers is placed. This would fulfill two functions: to fill the voids and to provide negative buoyancy against uplift if the trenches become saturated. Other types of backfill materials under consideration include concrete and bentonite mixes. However, it is also understood that other types of low permeability backfill materials would be considered for a given site-specific application.

The only form of actual external engineered barrier under active consideration is the cover unit. Covers would consist of layers of low permeability materials, combined with drainage layers. Most of the proposed designs incorporate a concrete slab above the disposal trenches and boreholes, which would have the following functions:

a) to support the overlying earth cover;
b) to serve as an intrusion barrier;
c) to inhibit water infiltration; and,
d) to retard migration of radionuclides.

The hydraulic barrier component is indicated to consist of clay or till. The biological barrier component would consist of a layer of cobbles sandwiched between layers of granular filter materials, which would also act as drainage layers.

It is generally assumed that the LLW trenches would be founded at a depth of approximately 8 m. The disposal unit would be founded at a depth of 6 m and the cover layer would be 2 m thick. It is assumed that the depth of weathering fissures is 2 m and that it is acceptable to bury the LLW below this depth.

TABLE 2: OVERVIEW OF INTERNATIONAL LOW-LEVEL WASTE DISPOSAL PRACTICES

COUNTRY	HOST MEDIA	RANGE OF DISPOSAL DEPTH	BARRIER TYPES		PROCESSING METHODS	WASTE ACCEPTANCE CRITERIA	WASTE PACKAGE Q.A. SYSTEM	COMMENTS
			INTERNAL	EXTERNAL				
U.S.A.	soil	shallow	• concrete • bitumen	• concrete container • steel container • soil backfill	• incineration and ash solidification	Yes - well developed	- - -	
SWEDEN	• crystalline rock below seabed (Forsmark) • soil (at 2 reactor sites only)	min. 50m rock cover	• cement • bitumen	• concrete container • steel container • clay liner (silos) • concrete walls (some caverns)	• incineration and ash solidification • compaction	Yes - very well developed	Yes - well developed	• Interim storage in surface facilities. • Backfilling in repository not intended.
FINLAND	• bedrock repository	deep	• cement • bitumen	• cement encapsulation • steel drum	• drying and solidification (ion exchange resins)	Yes	Yes - developing	• At present interim storage is in 300 m^3 concrete silos on surface. Therefore, less pressure to complete final repository.
WEST GERMANY (FRG)	• abandoned iron ore mine (KONRAD) • salt dome (Gorleben)	deep	• cement	• concrete container • steel container	• incineration and ash solidification • compaction • cutting • drying • sludge solidification	Yes - very well developed	Yes	• Backfilling in repository not intended. • May consider shallow burial in the future.
SWITZERLAND	• marl, anhydrite, or granite formations	deep	- - - -	• concrete container • steel container • plastic cement backfill	- - -	Yes - very well developed	Yes	• Emphasis is on host media barrier.
BELGIUM	• 100 m thick clay stratum	deep (200 m)	• high temp. incineratio (HTSI) - for HLW & TRU-waste but may be used for ILW & LLW	• bitumen • organic resin	• encapsulation	Yes	Yes	
NETHERLANDS	• to be decided	to be decided	• none at pre	• galvanized steel drum • concrete container or overpack	• compaction		- - -	• At present interim storage is at a surface facility near Petten.
ITALY	• soil	shallow	• cement if necessary	- - -	• solidification - cement	Yes - well developed	Yes - developing	• At present interim surface storage facilities are used.

NOTES: --- Data not obtained.
Information in this table extracted from reference 7.

TABLE 3: LOW-LEVEL WASTE DISPOSAL BY AECL - CRNL SITE PROPOSED ENGINEERED BARRIERS

HOST MEDIA	DEPTH	BARRIER TYPES	COMMENTS
Sand	10 m	Waste Form:	
		. bitumen	Waste form to be well characterized with regard to activity contact
		. cement, compressed waste	As overpacks
		. concrete	As overpacks
		. metals	
		Backfill/Buffer:	
		sand	
		mixed sand-clay	Placed in bottom of unit and as backfill
		clinoptilolite and sand	
		clays - illite, Na bentonite, kaolin, attapulgus	
		Covers and Walls:	
		reinforced concrete	May have coatings of bitumen, epoxy, or plastic membrane
		asphaltic concrete	
		plastic membranes (e.g., PE and hypalon) soil layers (clay, sand, gravel, cobbles, boulders), geotextile filters	

References: 3, 8, and 9.

TABLE 4: LOW-LEVEL WASTE DISPOSAL BY ONTARIO HYDRO PROPOSED ENGINEERED BARRIERS

HOST MEDIA	DEPTH	BARRIER TYPES	COMMENTS
Clay and (clayey) till	8 m or deeper	Backfill: Concrete, K-crete bentonite mixes, clay backfill Covers: layers of clay, concrete and drainage layers (sand and gravel)	
Limestone and Shale	up to 100 m	K-crete plug Backfill: clay till or bentonite mixtures	propose dry sand-bentonite to fill voids in ILW

References: 11, 12 and 13.

4. PROPERTIES OF ENGINEERED BARRIER MATERIALS UNDER CONSIDERATION

Engineered barriers (or liners) are commonly used for containing liquid pollutants, which are produced from municipal and industrial processes. Barriers perform one or both of the following functions:

a) To inhibit movement of liquids into the subsoil through the low permeability barrier material; and,

b) Absorb and adsorb pollutants as they move through the barrier material.

The types of barriers generally in use are summarized in Table 5. The barrier materials currently under consideration by AECL and Ontario Hydro are identified in the table.

4.1 Compacted Clay Barriers

Historically, compacted clay barriers have been used extensively for liquid waste impoundment. The soil material should have as low a permeability as possible and a mineralogy and chemistry that contribute to a high adsoprtion capacity.

The range of hydraulic conductivity (permeability) of compacted clay barriers is generally between 10^{-8} (illite) and 10^{-11} m/s (montmorillonite). To attain the low end of this range, it is desirable that the clay fraction (passing 25 microns) be as high as possible, preferably in excess of 25 percent.

4.1.1 Clay Chemistry and Mineralogy

Clay minerals have the advantage of being able to absorb ions, particularly cations, which may be present in a contaminant liquid. Hence, the common use of clays in engineered barriers.

A number of factors influence the behaviour of clay soil and, in particular, its performance when used as a barrier in contact with waste leachate. Fundamental factors affecting the performance of a barrier composed of natural materials are as follows:

a) clay content
b) clay mineralogy
c) cation exchange capacity (CEC)
d) soil permeability
e) pore water chemistry

The fundamental physico-chemical properties of clay minerals and their influence on the behaviour of compacted materials are discussed as follows:

Clay Mineral Content: The percentage of clay minerals present in the soil is a major factor controlling the material's volume change behaviour and hence its hydraulic conductivity. A clay mineral is defined as a crystalline material that exhibits plasticity because of its inherent ability to retain water.

Cation Exchange: The surfaces of clay minerals are negatively charged. As a result, cations such as K, Na, Ca and Mg present in the water in the void space are attracted to the clay particles in order to balance the negative charge and keep the whole unit electrically neutral. These cations are not held strongly to the particles and they

may be replaced or exchanged by other cations. Cation exchange refers to the process whereby attracted (adsorbed) cations are replaced by other cations.

The most significant aspects of behaviour of the three principal clay mineral components are as follows:

a) Kaolinite

Kaolinite has a relatively small surface area and tends to be chemically inactive, displaying little tendency for isomorphous substitution. As a result, kaolinite tends to be insensitive to changes in moisture, making it resistant to shrinkage and swelling. In this respect, it tends to behave much like fine silt.

b) Illite

Illite particles tend to be well bonded by potassium ions and hence, water penetration between the interstitial spaces is impeded, thus limiting the amount of swell. Although it has a relatively high negative charge, its real cation exchange capacity (CEC) is not very high, being intermediate between that of kaolinite and smectite.

c) Smectite

Smectite, which is the main clay mineral component of bentonite, has the smallest particle sizes and the greatest swelling potential of the three primary clay minerals. Attractive charges between adjacent layers are relatively small and smectite thus

absorbs polar organics and inorganic ions. It has a high surface area, cation exchange capacity and potential for water absorption. It also has the greatest capacity for shrinkage if the water is displaced by other liquids having a chemistry tending to reduce the interlayer spacing. When divalent cations (Ca^{2+}, Mg^{2+}) are present, smectite tends to disperse, with a corresponding higher permeability.

Sodium montmorillonite, a smectite mineral, tends to adsorb more water than other clay minerals, which generally results in lower permeability. This tendency to increased swelling makes it more susceptible to dispersion and erosion, as well as strength loss. Only sodium-rich montmorillonite is usually utilized for barrier materials, such as in admix liners, because of the relatively low permeability.

4.1.2 Clay Attenuation Capacity

The capacity of clay material to absorb the common ions such as Na, K, Ca and Mg, has been reasonably well researched. However, the absorption capacity of other ions associated with toxic waste compounds is poorly understood. Some of the difficulties with simulating field transport of solutes are as follows:

a) Most of the flow probably occurs as unsaturated flow, whereas most laboratory studies address saturated flow.

b) The chemistry of toxic wastes is very complex and, hence, difficult to model.

c) Field monitoring of trace contaminants has been sparse and hence, field data are lacking.

Another problem, which is discussed by Fuller[14], is that a poor correlation is observed between the attraction of toxic ions (such as Cd, Pb, and As) and the cation exchange capacity (CEC). Other mechanisms appear to control ion adsorption and hence the attenuation capacity of the soils. It is generally concluded that the attenuation of a particular soil must be studied for the particular wastes under consideration for containment.

4.1.3 Organics

Organic compounds can have highly detrimental effects on swell and permeability characteristics of clays. Organic molecules are adsorbed onto clay surfaces either through the vapor phase, or from aqueous solutions. The adsorption process is complex and involves a series of interactions between the organic molecules and the clay surface.

The basic mechanisms responsible for formation of clay organic complexes can include:

a) Ion Exchange: Organic ions will be adsorbed at clay mineral surfaces by ion exchange to neutralize the charges responsible for the cation (or anion) exchange capacity of the minerals.
b) Hydrogen bonding.
c) Covalent bonding.
d) Ion-dipole and coordination.
e) van der Waals forces.

Further information on clay organic complexes is contained in Yong[15].

4.1.4 Discussion

Waste leachate can affect the value of hydraulic conductivity of clay soils used in a barrier in several ways. Because of the complex nature of the waste, it may be difficult to predict whether exposure to the waste will increase or decrease the clay conductivity. However, several interactions can potentially increase conductivity, including:

a) Ion exchanges in which soil fabric is made more porous;

b) Piping of clay particles with reduced double layers through a predominantly gramular matrix;

c) Dissolution of soil minerals by acids and bases;

d) Syneresis of highly polar organic molecules; and,

e) Dissolution of clay minerals by complexation.

A laboratory testing program should be prepared for the actual waste, to determine the effects of key chemical constituents of the permeant on the soil barrier[16], including permeability and sorbent properties.

4.2 Concrete and Cement

Concrete is a mixture of Portland cement, fine aggregate, coarse aggregates, air and water. It is a temporarily plastic material, which can be cast or molded, but is later converted to a solid mass by chemical reaction. Concrete is formed by a complex series of hydration reactions. The principal reaction within the cement is the hydration of calcium silicates to calcium silicate hydrate and calcium hydroxide. Initially, the calcium silicate hydrate forms a gel which develops a

rudimentary crystalline lattice, giving strength and rigidity to the concrete. The hydration reaction is exothermic, producing relatively high temperatures after mixing. This can induce thermal stress and cracking within the concrete.

Portland Cement Type 1 (CSA Type 10) with or without fine aggregate, is commonly used for the encapsulation of radioactive wastes. This is due to its satisfactory physical properties, general availability, capability of providing a freestanding monolith and waste stability during subsequent handling.

Additives may be mixed as ingredients in the concrete or cement waste immediately before or during mixing. Many additives are available to modify, improve, or give special properties to concrete or cement waste mixtures. Commonly used additives are:

a) Air entraining agents to increase the resistance to frost action, thereby improving durability. Agents in most frequent use are natural and synthetic soaps.

b) Pozzolans to reduce the heat of hydration, increase resistance to sulphates and prevention of calcium hydroxide leaching. Pozzolans are siliceous substances, such as fly ash and pumice, that react with lime in the presence of water. Disadvantages are increased shrinkage with drying and reduced durability.

The more important admixtures being investigated are silica fume, blast furnace slag (BFS), fly ash and clinoptilolite. Typical cements and admixtures presented being considered are shown in Table 6. Chemical composition for more important admixtures is presented in Table 7.

On addition to concrete and cement, silica fume reacts rapidly with hydration product calcium hydroxide to form a compact form of calcium silicate hydrate. This reaction results in a rapid increase in strength with a corresponding significant decrease in the permeability. Fly ash and blast furnace slag have similar reaction mechanisms to silica fume, but the rate of reaction is much slower due to the smaller percentage of silicon dioxide. Other hydration reactions do occur with fly ash and blast furnace slag due to the presence of lime and aluminium oxide.

Clinoptilolite is being considered by CRNL as an internal buffer material, but also can be used as an admixture to cement and concrete. This naturally occurring mineral is used in nuclear applications for waste water treatment on account of the high specificity for Cesium. Its cation exchange capacity is in the region of 200 - 230 $mg.Cs.g^{-1}$. Water permeability for these blended concretes and cements ranged from 10^{-11} to 10^{-16} m/sec, the lower values being obtained by the addition of silica fume. Gas permeability values tend to be at least two orders of magnitude higher.

One additive under investigation for waste encapsulation[17] is blast furnace slag, which possesses an ameliorating influence on the hydration reactions. The addition of blast furnace slag moderates the hydration reactions so as to give a lower temperature and less thermal stress.

4.3 Bentonite and Sand Admixes

Bentonite and sand admixes consist of powdered, or granular bentonite, mixed with native sand in ratios that result in a low permeability

material, after the bentonite has hydrated and swollen to plug the voids in the sand. High swelling sodium bentonite (i.e., where sodium is the predominant exchangeable cation in the bentonite clay structure) is generally preferred. The bentonite application rate required to produce a liner hydraulic conductivity that meets the specified seepage control criterion should be determined by laboratory permeability tests.

4.4 Asphalt and Asphalt Admixes

Asphalt is the black or dark brown derivative from petroleum evaporation and, for the purpose of this project, is considered synonymous with bitumen. Asphaltic materials used in barrier systems include hydraulic asphalt concrete (HAC), spray-on bitumen membranes and soil asphalt admixes. The only asphalt liners generally in use in non-toxic liquid containment are:

a) spray-on bitumen over HAC; and,
b) spray-on bitumen over soil asphalt.

Asphalt is generally resistant to most acids, bases, organic salts and some organic materials. However, it is attacked by organic solvents, particularly hydrocarbons. Therefore, HAC would not be suitable for containing waste with a significant component of petroleum substances.

Asphalt admixes provide support for relatively weak superimposed membranes, while bituminous membranes provide additional seepage control over the more crack-susceptible HAC, soil asphalt and concrete covers. HAC generally requires an asphalt content of about 7 to 9% and a porosity below 4% for suitably low hydraulic conductivities.

Soil asphalt is a mixture of a silty gravel soil and liquid asphalt. This material, unlike bitumen, is not usually of sufficiently low permeability, and therefore, requires a water-proof seal[18].

4.5 Soil Cement

Soil cement is a mixture of Portland cement, water and various natural soils. Soil cement mixtures have been used in recent years in construction of dams as a substitute for concrete and compacted fill materials. The soil cement mixtures are generally strong, although the permeabilities are relatively high (10^{-8} m/s)[19]. To achieve a lower permeability, coatings, such as epoxy asphalt can be used.

Suitable soils for use in cement mixtures would have less than 50 percent by weight of silt and clay fines. A well graded mixture ranging from clay to fine gravel is the preferred material, to maximize the density of the mixture. Acidic environments can result in deterioration of the cement. However, soil-cements are generally resistant to alkalis and organic compounds. One of the main disadvantages of soil-cement in northern climates is potential cracking due to freeze-thaw effects, as well as crack development associated with shrinkage during drying.

4.6 Polymeric Membranes

Polymeric membranes are being proposed by AECL as part of the cover system to limit infiltration into the low-level waste disposal unit. Considerable experience has been gained concerning the use of polymeric materials in liners for the retention of liquid wastes. A comprehensive review of the current state of the art of synthetic liners is presented by Landreth[20] and performance aspects are discussed in Section 5.4.

Polymeric membranes are synthetic plastic or rubber sheets that are seamed in the field using solvents, adhesives, or welding processes to form continuous membrane liners. There are several polymers and resultant compounds that are made into liner sheets, and these have a wide range of material properties. The most common polymeric liner materials

in use are polyvinyl chloride (PVC), chlorinated polyethylene (CPE), chloro-sulphonated polyethylene (Hypalon*) and high density polyethylene (HDPE). Membrane thicknesses range from about 15 mils** for PVC up to 100 mils for some HDPE liners. The hydraulic conductivity of intact membranes is generally not measurable, although membranes are susceptible to punctures and faulty seams if strict quality control is not exercised.

Many polymers may be produced in vulcanized (elastomeric) or unvulcanized (thermoplastic) form. Vulcanized liners may be stronger and more chemically resistant, but thermoplastic versions of compounds like chlorinated polyethylene (CPE) and Hypalon are more common. Thermoplastics are easier to seam and repair in the field. Table 8 shows several types of polymeric liners and their availability as vulcanized or thermoplastic material.

4.7 Metals

Where there is little ingress of water, a reducing environment will develop in the respository and metals will tend to be stable. However, if groundwater is replenished at a sufficiently high rate, oxygen will tend to gradually cause oxidation. If groundwater is present, it is essential to select a metal that corrodes very slowly and to specify a sufficient thickness so that it will not fail within the desired life of the barrier. Failure occurs usually by localized corrosive attack such as pitting, in crevices, stress corrosion cracking and intergranular attack.

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* Hypalon is a registered trade-mark of DuPont, and is available from several liner manufacturers and suppliers.

** A mil is equal to 0.001 inch and is the common unit for describing membrane thickness. A thickness of about 39 mils is equal to 1 mm.

Various metals have been evaluated[21,22] for use in canisters and overpacks for containing radioactive waste. Materials were placed in an environment where water was relatively aggressive and radiolysis was a source of oxygen replenishment. The canister materials studied were: carbon steel, stainless steel, inconels, aluminum, monel, copper-nickel, copper and three types of titanium. As the carbon steel is significantly less costly than the other metals, it received the most attention. The stainless steel was of interest because with its low carbon content; it can be welded without increasing its susceptibility to corrosion. The inconel alloys are nickel-based and very resistant to corrosion. Tests were conducted at 100 and 300 C. Conclusions reached were as follows:

a) Stainless Steel - susceptibility to chloride induced corrosion; generally susceptible to pitting, crevice corrosion and stress corrosion. Not recommended if chlorides present.

b) Iron and Low Alloy Steels - poor resistance to aqueous solutions, the severity of attack increasing with salt content. Recommended only if kept dry.

c) Aluminum Alloys - susceptible to pitting and stress corrosion cracking. Presence of heavy metal ions in solution greatly increases pitting. Not recommended for canisters.

d) Nickel Alloys - potentially acceptable, but requires long term confirmatory testing.

e) Zirconium Alloys - highly resistant to aqueous salt solutions and most acids and bases. Subject to stress corrosion cracking in chloride solutions.

f) Titanium Alloys - very resistant to chloride salt solutions, but more susceptible to attack by acids and bases. Probably most resistant metal under consideration.

Long term corrosion rates are very difficult to predict from short term tests and with only limited knowledge of the chemistry of the waste material and groundwater and mechanics of corrosion. The preliminary review of limited information on metals suggests that none of the metals considered will remain completely unbreached for even 50 or 100 years. Also, in view of the high cost of metal barriers, it appears unlikely that they would be considered as a cost effective engineered barrier for use in low-level waste containment.

TABLE 5.

COMMONLY USED BARRIER MATERIALS

CLASS	TYPICAL MATERIALS	UNDER CONSIDERATION BY AECL AND/OR ONTARIO HYDRO (Y = Yes)	PERMEABILITY OF INTACT MEMBRANE (m/s)	REMARKS
Concrete	Cement, coarse and fine aggregates, fly ash additives (K-crete)	Y	10^{-10} - 10^{-12} 10^{-10} - 10^{-15}	
Compacted fine-grained soil	Local clayey soil Clays - illite, bentonite, kaolinite, attapulgus	Y Y	10^{-8} - 10^{-10}	Porous, discontinuous, economical, typically 0.3 - 1.2 m thick.
Admixes	Bentonite or sand-bentonite Soil cement Asphalt and soil-asphalt (Hydraulic) asphatic concrete (HAC)	Y Y Y Y	10^{-7} 10^{-7} 10^{-8} 10^{-9}	Low permeability binder mixed with native soil, except for bentonite may be relatively rigid.
Polymeric membranes: Thermoplastics	Polyvinyl chloride (PVC) Chlorinated polyethylene (CPE) Chlorosulphonated polyethylene (CSPE) (Hypalon)* Elasticized polyolefin (ELPO)	Y	10^{-12}	Continuous liner, relatively expensive, typically 0.5 - 2.6 mm thick, may be reinforced with polyester scrim.
Vulcanized elastometers	Butyl rubber Neoprene (CR) Ethylene propylene diene monomer (EPDM)		10^{-12}	May be partially pre-assembled.
Crystalline thermplastics	Low density polyethylene (LDPE) High density polyethylene (HDPE)	Y	10^{-12}	Diffusion barrier for tritiated wastes.
Spray-ons	Catalytically blown asphalt Emulsified asphalt Air blown concrete (shotcrete)	Y	10^{-8}	Continuous liner, discontinuous at pinholes, cracks, typically 4 - 8 mm thick.
Sealants	Polyacrylamide Liquid vinyl polymer		10^{-7}	Sprayed, dusted or ponded, may result in nonuniform coverage.
Chemisorptives			n/a	Function is to absorb contaminants, experimental.
Metals	Steel, aluminum	Y	negligible	

* Registered trademark of DuPont.

TABLE 6: A LIST OF THE CEMENT-BASED INTERMEDIATE AND LOW-LEVEL ENCAPSULATION MEDIA UNDER REVIEW (from Rushbrook[17])

	CEMENT CONSTITUENT	ADDITIVES
1.	Ordinary Portland Cement (OPC)	-
		Clays (vermiculite), zeolites, slicia, diatomaceous earths
		Pulverized Fuel Ash
		Blast Furnace Slag
		Slag or ash and clays
		Sodium Silicate
		Incorporating polymers*
		Impregnated with polymers*
		Silica fume
2.	Sulphate Resistant Cement	Blast Furnace Slag
3.	High Alumina Cement	Clays (nontronite, smectite, illite, tobermorite, gibbsite)
		Pozzolana
		Silica fumes
4.	Light Weight Cement	Vinsol - Resin, air entrainment.

* Notably epoxy and polyester resins.

TABLE 7: CHEMICAL COMPOSITION AND PHYSICAL CHARACTERISTICS OF THE VARIOUS ADMIXTURES (Gjorv[74])

		BLAST FURNACE SLAG	CONDENSED SILICA FUME	FLY ASH
SiO_2,	%	31.7	93.1	41.0
CaO,	%	39.3	0.2	3.1
Al_2O_3,	%	6.7	0.3	22.8
MgO,	%	13.3	0.6	0.8
Fe_2O_3,	%	0.5	0.6	18.2
L.O.I.,	%	0.9	3.5	9.5
TOTAL		92.4%	98.3	95.4
Blaine m^2/kg		390	19 500 (B.E.T.)	-
Specific Gravity		2.90	2.20	2.42

TABLE 8: POLYMERIC MATERIALS USED IN MEMBRANES

POLYMER	AVAILABLE AS		AVAILABLE AS	
	THERMOPLASTIC	VULCANIZED	REINFORCED	UNSUPPORTED
Butyl rubber	NA	X	X	X
Chlorinated polyethylene	X	X	X	X
Hypalon	X	X	X	NR
Ethylene propylene rubber	X	X	X	X
Neoprene	NA	X	X	X
High density polyethylene	X	NA	NA	X
Polyvinyl chloride	X	NA	X	X

NOTE: X indicates available.
NR indicates not recommended.

5. REPORTED PERFORMANCE OF ENGINEERED BARRIERS IN STORAGE OF INDUSTRIAL AND MUNICIPAL WASTES

The performance of a particular barrier material in waste containment depends on the actual leachates which contact the barrier during its lifetime. Leachate composition can change with time and is generally very complex. To minimize potential effects, as much information as possible should be known about the composition of the waste during the lifetime of the disposal facility. No single type of barrier material is likely to be capable of resisting all waste types.

Industrial waste compounds can typically include highly polar solvents (water), highly non-polar material (oils and hydrocarbon solvents) and a wide range of other pollutants. Dissolved organic compounds in the leachate can cause the swelling, strength loss and eventual failure of many synthetic liner materials which are based on organic compounds, which are primarily polymeric materials. Organic liquids can also exercise dramatic changes on the permeability of some clays (see Section 5.1.1).

The U.S. EPA, recognizing the lack of information concerning the effects of different chemicals on barrier materials, has sponsored several research programs[23,24,25,26,27].

The performance of the different types of barrier materials under consideration for a variety of waste service environments is discussed under the respective material types in the following sections. The qualitative performance of several industrial wastes, in contact with various engineered barriers under consideration, is shown in Table 9.

5.1 Clay Barriers

This section applies both to clays used as engineered barriers and which constitute the surrounding host media. Potential effects of interactions between waste leachate and clay materials are as follows:

a) Volume Change: The clay may either shrink or swell if water is replaced by leachate. The water absorption potential is controlled by the dominant cation in the liquid. For instance, the Na-cation results in high absorption of water in smectites. By contrast, basic aromatic compounds, such as analine, can result in expulsion of water and shrinkage.

b) Electrical Properties of the Liquid: Organic compounds tend to result in a lower dielectric constant, which contributes to a smaller interlayer spacing and increased permeability. To determine the effect of a replacement liquid on permeability, it is generally necessary to conduct permeability tests using the pollutant as the test fluid.

c) Chemical Dissolution of the Clay Minerals: Some acids and bases can dissolve aluminum, iron and alkalies, as well as silica. These are all components of the clay minerals. The process of dissolution is referred to as acidization, which results in an increased permeability. Organic acids may be present in waste material because of the decomposition of waste anaerobic decomposition. Typical liquid decomposition byproducts are propionic, butyric, isobutyric and lactic acids[18].

d) Piping Erosion: This is usually a problem only where the waste is completely liquid and subjected to relatively large hydraulic pressures. Piping is not expected to be a failure mechanism at a solid waste site.

5.1.1 Effect of Organic Liquids on Permeability

Various studies have been conducted concerning the effect of different permeants on soil permeability. It has been found that some organic chemicals can have a dramatic effect on increasing the permeability of clayey soil. It is generally recommended to test the permeability of potential clay lining material with the actual organic liquid that would be expected to permeate the soil.

Industrial liquids of low dielectric constant may produce very large increases in the permeability of a water saturated clay barrier if mutually soluble liquids such as acetone, glycol or alcohol are present. The presence of a concentrated zone of such liquid waste over a clay liner could conceivably create a significant, highly conductive pocket in the barrier.

Quigley et al[28] reported that soils mixed with pure liquid hydrocarbons of very low dielectric constant can have permeabilities of up to four to six orders of magnitude higher than those obtained when water is the pore fluid. However, in a field situation, the clay would be water saturated and it may be difficult, though not impossible, for a hydrophobic contaminant to penetrate a water-saturated barrier. Even if such a liquid were forced into the clay barrier, indications are that it would not react with the clay and the permeability would not necessarily increase.

Permeation of the clay barrier by polar organic compounds soluble in water may increase hydraulic conductivity by 5 to 10 times. Industrial waste mixtures with abundant alcohol present as an association liquid between polar water and non polar aromatics may very severely damage a clay barrier, although research into these interactions is incomplete.

Other studies[29] have been done on the effect of various liquids on clay permeability, with the general conclusion being reached that permeabilities can be increased by up to 2 orders of magnitude, or more. There is no simple correlation, however, between permeability and organic content and in some cases, permeabilities have even been found to have decreased with certain substances. The need for specific permeability testing using the actual liquids likely to be present in the waste leachate is therefore reinforced.

5.1.2 Effect of Inorganic Substances on Permeability

In general, it has been found that when the pore fluid anionic and cationic concentration is increased, the soil permeability is reduced by an amount which is proportional to the sodium absorption ratio (SAR). Nevertheless, exceptions do exist. In the case of a montmorillonitic soil, which was tested[30], this trend was found to be reversed. For a low SAR, the permeability did drop with decreasing salt concentration. However, at increasing salt concentration, the trend was reversed. This would indicate that a lining material with a high percentage of montmorillonite could undergo an increase in permeability if the waste effluent is more concentrated than the pore solution in the liner. This effect would be undesirable and hence some caution should be exercised in the use of high montmorillonite clay liners.

Clay covers having a permeability lower than about 10^{-8} cm/s are reported to be effective in radon attenuation[31].

In compatibility tests for acid wastes, EPA found that the permeability of most soil liners was relatively low, being in the order of 2 x 10^{-10} cm/s. Fluids seeping through the soils were found to be essentially neutral (pH 6-8), but with a high dissolved solids (salt) content. There was also a tendency for the solids content to reduce with time, except for caustic waste. Except for the liner exposed to the caustic waste, the pH of the liners was not significantly altered by the wastes.

5.2 Concrete

The use of high-integrity reinforced concrete containers for disposal of radioactive wastes and specifically EPICOR prefilters was reported by Schmitt and Reno[32]. A test procedure has been developed under DOE Auspices to ensure the satisfactory performance of concrete containers. For example, the container may be dropped 25 feet onto a rigid surface to demonstrate its robustness. The Nuclear Regulatory Commission has indicated that a high-integrity container should have a minimum life of 300 years, resist corrosive and chemical effects of the waste and surrounding geological materials, resist biodegradation and provide a positive seal throughout the design life[32].

Although concrete is relatively durable, it is prone to eventual cracking for several reasons, including frost action settlement, as discussed by Trevorrow et al[33].

5.3 Soil Cement and Grouts

In the testing program conducted by Matrecon[27], various industrial waste types, except acid, were tested on soil cement. No seepage was observed over 2.5 years of exposure. An increase in compressive strength was observed during the period of testing. Minor blistering of the epoxy asphalt coating was reported. The tests provide good evidence of at least the short term suitability of soil cement for containing hazardous waste leachate.

In exposure tests to municipal solid waste (MSW) leachate, soil-cement mixture was observed to undergo normal hardening (hydration) process with time, although some loss in strength was observed. The permeability decreased over the period of exposure. It was suggested that inhomogeneities in the admix material indicated the need for a much thicker liner than might otherwise be theoretically necessary.

Cement-clay grouts, as well as polymeric and bituminous grouts, were assessed for their compatibility with different classes of organic and inorganic chemicals, which may be present in industrial wastes, as reported by Hunt et al[34]. Interactions between the grouts considered and different chemical groups are summarized in Table 10. Field testing was conducted at one LLW disposal site, where a cement-bentonite grout indicates promise as a barrier[35]. Information on grout/chemical compatibility is quite limited and is probably insufficient for design, thus necessitating laboratory testing with the actual chemicals anticipated to come into contact with the grout material proposed for use in the barrier.

5.4 Polymeric Membranes

Both organic and inorganic substances can result in chemical attack of liners. The most serious effect is swelling of the polymeric material, which can lead to softening and eventual failure. Also, some of the

materials which contain large amounts of monomeric plasticizer (e.g., PVC) may lose plasticizer and, as a result, will tend to shrink and break.

Matrecon Inc.[27] conducted only two field and laboratory studies of the effects of hazardous liquids on polymeric and other types of linings. Hence, only limited data are available concerning the effects, particularly in the long term, of waste liquids on polymeric membrane liners. Much of the evidence indicating the likely effects is based on laboratory experiments. Most of the documented field cases relate to the impoundment of water and waste liquids, although in recent years there have been some trials concerning solid waste containment (MSW leachate). The use of polymer liners in waste disposal sites began in the early 1970's. Liner types that were tested are as follows:

- Butyl rubber
- Chlorinated polyethylene (CPE)
- Chlorosulphonated polyethylene (CSPE)
- Ethylene Propylene rubber (EPDM)
- Polyvinyl Chloride (PVC)

Each of these materials was exposed to MSW leachate in laboratory submersion tests. The liner condition was determined after 12 and 56 months of exposure to the leachate. It was found that, except for imperfections in some of the sealing, none of the liners exhibited any seepage. However, a substantial reduction of seam strength was found for the CPE, CSPE, and EPDM materials. The reduction in seam strength for the CPE dropped from 57.0 pounds per inch to 17.0 ppi after 56 months; the CSPE dropped from 50.0 to 10.0 ppi after 56 months. If the seams had been subjected to stress, it is possible that rupture could have occurred. Other than for the reduction in seam strength, none of

the membranes exhibited significant changes in properties. It is considered that the seam deterioration could be compensated by improvements in seaming techniques and quality assurance that have been developed in recent years.

In another EPA study, polymeric liners were exposed to organic acids containing acetic propionic, isobutyric and butyric compounds (concentration of 21.9 g/L)[24]. The absorption of the lechate containing the organic acids was compared with the absorption of plain water. Two of the membranes testes (PVC and EPDM) exhibited relatively large increases in the percentage of leachate absorbed by weight after a one year exposure period. The EPDM absorption increased from an average of 2.3 percent in water to 6.6 percent in the organic leachate; the PVC increased from 1.65 to 4.1 percent. Some of the other liner materials tested (CSPE, CPE and neoprene) exhibited a larger absorption of water (e.g. average of 16 percent for CPE), but less of the organic leachate was absorbed (CPE dropped to 10.6 percent), indicating that the individual polymers react differently under exposure to specific organic compounds contained in the leachate. No drastic deterioration in properties was observed. In general, in the testing; however, it should be noted that the duration of testing was only for one year which may be too short a period to be indicative of long-term trends. Also, the total composition of the leachate and the concentration of organic compounds will affect the percentage absorption of the leachate.

Only a few case histories of the field effects of MSW on polymer liners have been reported, as described by Matrecon Inc.[27]. These involved four abandoned disposal sites as follows:

a) A 30 mil PVC liner was in service for six years at an Ohio landfill. Little overall change in properties was reported, although the placement of a clay liner over the PVC may have separated the membrane from coming into direct contact with the MSW leachate.

b) A 15 mil PVC liner was in service at a sludge lagoon for 6.5 years. Where the liner was covered with either soil or sludge, deterioration was negligible. However, portions of the sheet which had been exposed to weathering, for much of the exposure period, had become very brittle and decomposed. For example, the seam tear strength decreased from in excess of 20 pounds per inch, where the material was covered, to practically nil where it was exposed. The authors[19] conclude from the observations that a PVC liner must be covered and should be more than 15 mils thick. However, it was also observed that even some of the buried liner had undergone a loss of plasticizer, indicating that even the buried membrane may have undergone long term deterioration.

c) At another site, three different types of lining - CSPE, CPE and LDPE - were tested for nine years under the direction of the Solid and Hazardous Research Division of EPA[36]. The MSW leachate in contact with the materials was considered to be relatively dilute, because of a layer of clay soil placed on top of the liner. The CSPE liner swelled by up to approximately 28%. The LDPE showed little swelling or loss of physical properties. However, the CPE lining absorbed significant leachate and became "stiff and leathery". The maximum swell was estimated to be approximately 32%. Although the CPE material had swollen significantly, its physical properties remained acceptable, at least over the duration of the trial.

d) At a pilot scale landfill at the Georgia Institute of Technology, a CSPE liner was tested for a 4 year period. The material exhibited some swell, but no major change in properties was reported.

A laboratory study of exposure of a range of lining materials to a variety of hazardous wastes was conducted under EPA auspices[21]. Wastes used in the tests included two strong acids, a strong alkali waste, oil refinery tank bottom sludge, a lead waste from leaded gasoline, saturated and unsaturated hydrocarbons, and pesticide. Specimens were tested after 1 year and 3.5 years of exposure. A total of eight different membranes were tested as follows: Butyl, CPE, CSPE, ELPO (elasticized polyolefin), EPDM, neoprene, polyester and PVC.

The polyester elastomer lost its elongation upon exposure to strong nitric acid. The CSPE, neoprene and EPDM materials had a significant reduction in elongation in the alkaline, lead and nitric acid wastes. The CSPE and PVC stiffened and the neoprene softened in exposure to these wastes. Immersion tests were done in conjunction with the laboratory seepage tests. Eight different polymers (a total of 12 membranes) were tested in eight different wastes for two years. One of the oily wastes, a weed oil, caused substantial swelling in the CPE and CSPE liners. A naptha oil had a significant effect on the butyl rubber. It was also found that among the three PVC specimens tested, swell and elongation effects were quite different. From pouch tests, it was found that the PVC lining was usually more permeable than the other types.

It was found, in general, that where the liners were exposed directly to the waste, significant losses of properties did occur. However, where an intermediate soil was present as protection between the liquid and

the liner so that the waste was not in direct contact with the liner, losses were small. This raises the concept of a dual membrane-soil liner as being a potential optimum engineered barrier system.

Organic materials can be absorbed by organic lining materials (ashphalt and polymer membranes). In testing reported by Matrecon[28], a range of absorption of organic compounds was reported. The most resistant material to oil absorption was HDPE (less than 1%) to the highest gain in CPE (107%).

Conclusions reached in the above studies, as presented in the EPA report, include the following:

a) most polymeric materials will tend to swell somewhat.
b) cross linking of a polymer or rubber reduces the swell.
c) crystallinity of the polymer also reduces the tendency to swell and hence dissolve. The most resistant material is a highly crystalline polymer, such as HDPE, which will not dissolve in gasoline. This product is generally recognized as being the most, although not completely, effective material in lining sites subject to gasoline spills.
d) Soluble constituents present in polymers may increase the amount of swell, as a result of diffusion of water.

Swelling is therefore considered to be a reliable index for indicating the probable liner performance when exposed to a particular waste. It may be possible to correlate the probable lifetime of a membrane with the rate of swelling, although the authors are not aware that an acutal procedure has yet been developed for such a correlation.

Other failure mechanisms which must be considered in membrane designs are discussed in Section 8. These include possible effects of frost and gas, differential settlement causing elongation and stress cracking and biodegradation. Detrimental effects associated with swelling are as follows:

a) Strength loss and increased creep.
b) Elongation loss.
c) Increase in permeability, more rapid aging and polymer degradation.

5.5 Asphalt and Asphaltic Admixes

In exposure tests to MSW leachate, EPA[20] found that both asphalt concrete and soil asphalt liners exhibited a dramatic loss of strength after 1 year of exposure, although the permeability remained low. The bitumen binder appeared to absorb organic compounds.

Oily waste was not placed on bituminous material. However, lead waste, which contained light oily material, was examined for liner compatibility. Only the hydraulic asphalt concrete (HAC) was used in contact with the acid waste. Performance is summarized as follows:

HAC: Four types of waste were tested on this material, excluding oily wastes. Satisfactory performance was experienced under exposure to pesticide and caustic wastes; however, the liner failed to contain the nitric acid. This failure was attributed to leaching of the calcium carbonate aggregate.

The bitumen tended to absorb the oily component of the lead waste and became very soft.

Emulsified asphalt on nonwoven fabric: This composite membrane was exposed to pesticide, caustic and lead wastes. The acid waste was not tried because of anticipated excessive hardening. The oily waste was not tested because of high potential solubility of the asphalt in the oil.

Satisfactory containment was experienced with the pesticide and caustic wastes; however, significant seepage occurred from the lead waste.

5.6 Sand-Bentonite Admixes

EPA tested sand-bentonite mixes exposed to industrial wastes[20]. Measurable seepage occurred in 8 of 10 tests. It was concluded that this type of liner is probably not satisfactory for the waste types tested. Channeling tended to occur and seepage and incipient failure followed. No details were provided regarding the chemical composition of the leachate, or the test procedures and results obtained.

TABLE 9: QUALITATIVE PERFORMANCE OF BARRIER MATERIALS AND DIFFERENT CHEMICAL GROUPS (from Landreth[20])

LINER MATERIAL	CAUSTIC PETROLEUM SLUDGE	ACIDIC STEEL-PICKLING WASTE	ELECTRO-PLATING SLUDGE	TOXIC PESTICIDE FORMULA-TIONS	OIL REFINERY SLUDGE	TOXIC PHARMA-CEUTICAL WASTE	RUBBER AND PLASTIC
SOILS:							
Compacted clay soils	P	P	P	G	G	G	G
ADMIXES:							
Asphalt-concrete	F	F	F	F	P	F	G
Asphalt-membrane	F	F	F	F	P	F	G
Soil asphalt	F	P	P	F	P	F	G
Soil-bentonite	P	P	P	G	G	G	G
Soil cement	F	P	P	G	G	G	G
POLYMERIC MEMBRANES:							
Butyl rubber	G	G	G	F	P	F	G
Chlorinated polyethylene	G	F	F	F	P	F	G
Chlorosulfonated polyethylene	G	G	G	F	P	F	G
Ethylene propylene rubber	G	G	G	F	P	F	G
Polyethylene (low density)	G	F	F	G	F	G	G
Polyvinyl chloride	G	F	F	G	G	G	G

G - Good, F = Fair, P = Poor

TABLE 10: INTERACTIONS BETWEEN GROUTS AND GENERIC CHEMICAL CLASSES (adapted from Hunt et al[34])

CHEMICAL GROUP	GROUT TYPE											
		PORTLAND CEMENT					POLYMERS					
	Bitumen	Type I	Type II and V	Clay (Bentonite)	Clay-Cement	Silicate	Acrylamide	Phenolic	Urethane	Urea-formaldehyde	Epoxy	Polyester
Acid	?a	1d	1d	?c	?c	3a	2c	?a	2c	a	?a	?d
Base	?a	1d	1a	?c	?d	2c	3d	?d	?d	2c	?a	?d
Heavy Metals	?	1a	?	?d	?	3?	3?	?	?	?	?	?
Non-Polar Solvent	?d	2c	2?	?d	?	?	?a	?d	?a	2?	?d	?d
Polar Solvent	?d	2c	2?	?d	?	?	?	?d	?a	2?	?d	?d
Inorganic Salts	?d	2c	2a	2d	?d	3?	3d*	3a	?d	?a	?a	?

KEY: Compatibility Index

Effect on Set Time
1. No significant effect
2. Increase in set time (lengthen or prevent from setting)
3. Decrease in set time

Effect on Durability
a No significant effect
b Increase durability
c Decrease durability (destructive action begins within a short time period)
d Decrease durability (destructive action occurs over a long time period)

Except sulphates, which are ?c
Except KoH and NaOH, which are 1d
* Except heavy metal salts which are 2
Non-oxidizing
Modified bentonite is d
? Data unavailable

6. MICROBIOLOGICAL INTERACTION OF LOW-LEVEL WASTE FORMS AND ENGINEERED BARRIERS

6.1 Introduction

Microbiological activity has been shown in recent studies[37] to play an important role in the decomposition of municipal waste. In the United States, studies at low-level waste sites, such as Maxey Flats, have observed significant microbial activity. Supportive evidence for microbial interaction with waste forms is the detection of tritiated and Carbon-14 contaminated gases[38,39,40], and the presence of chelating agents[41,42]. These observations of radioactive releases may reflect poor installation and maintenance of the trench covers which have permitted entry of water.

Frissel et al[43] have summarized the mechanisms by which micro-organisms may affect the degradation of waste forms, the performance of engineered barrier materials and the mobility of radionuclides. These mechanisms include the following:

a) Direct utilization of the barrier materials and waste forms as either nutrient or energy sources. Radionuclides may become chemically altered via various biochemical pathways within the microbial cell and released in a different (may be more soluble) chemical form;

b) Production of organic or inorganic compounds (metabolites) which chemically degrade barrier materials and/or waste forms and/or solubilize radionuclides.

c) Alteration in environment of waste depository promoting increased microbial activity may result in an accelerated chemical deterioration of the barrier materials and waste forms.

In the forthcoming sections, the above processes will be discussed in the context of low-level radioactive waste forms and engineered barriers such as bitumens, epoxy resins, concretes, metals, plastics and soils (backfill) and the host geological media.

6.2 Microbial Activity Within Waste Repository (General)

This section addresses several aspects of microbiological activity including:

a) types of micro-organisms observed within shallow burial waste sites.

b) mechanisms or routes by which viable micro-organisms could be introduced into the waste depository.

c) general requirements for microbial growth and the impact of various environmental factors on growth.

d) conditions during construction and closure phases and their impact on microbial growth.

A review of the current literature on microbial aspects of low-level and intermediate-level waste disposal undertaken by Sharpe[44] and Rushbrook[17] concluded that micro-organisms are distributed throughout both sedimentary and crystalline host media to depths in excess of 400 m. Also studies undertaken by West et al[45] clearly indicate that micro-organisms are present in the groundwater.

Viable micro-organisms (bacteria and fungi) may be introduced into the waste forms or waste repository via various manners including:

a) contamination of the waste forms during temporary storage.

b) backfill and host geological materials inadvertently introduced during construction and closure phases.

c) micro-organisms associated with organic materials such as leaves and pollen (this will be discussed later).

d) groundwater intrusion from host media.

Examination of leachates removed from shallow land burial sites in the U.S.A. (Maxey Flats, Kentucky and West Valley, New York) showed the presence of populations of aerobic and anaerobic sulphate reducers, dentrifiers and methanogens. Although micro-organisms are present in almost all habitats including the groundwater, several environmental factors affect the rate of microbial activity. These include substrate, water, temperature, oxygen and Eh, pH and radiation[46]. Table 11 presents a summary of these factors and their impact on microbial growth.

With respect to a shallow burial repository, several distinct environmental conditions are likely to exist within the normal lifetime. During construction, the site would be aerobic with ambient temperatures prevailing. Providing rainwater or moisture enters the waste repository, microbial activity will be established if suitable substrates, as shown in Table 11, are available. Organic material not associated with the waste forms could be introduced into the repository either directly or inadvertently. Direct entry would come as leaves or pollen falling

from nearby vegetation, fecal matter dropped by birds or small animals, debris carried in by runoff or air movement, or left behind during construction or backfilling operations. If dry conditions do prevail within the repository, with a temperature cycling between 5 to 10 C, then microbial activity will be significantly reduced. Depending upon the microbial species activity an still be present (although markedly decreased) at temperatures below 5 .

6.3 Microbial-Waste Form Interaction

As commented in Section 2, the vast majority of low-level waste produced in Canada is solid waste. Excluding ion exchange resin (high activity) and liquid wastes, most of this radioactive waste has an activity of less than 1 Ci/m^3 and will be compacted into plastic wrapped bales prior to disposal. Low-level waste can be classified into three main types, i.e., plastics, paper and miscellaneous materials. Information compiled by Ontario Hydro, with respect to CANDU waste, indicates that plastic composed 15 to 62 percent of the waste, paper ranged from 16 to 45 percent and miscellaneous materials varied from 22 to 52 percent[5]. The miscellaneous materials include sweeping compounds, vermiculite, fibreglass insulation, metal pieces, electric cable, rubber boots, animal carcasses, food, gloves, cans, glass and plastic bottles containing oils and aqueous fluids. These waste materials provide some major nutrients for microbes in the categories of proteins, lipids, polysaccharides and aromatic compounds. This section will discuss specifically the interaction of the micro-organisms with the cellulosic and organic components of the waste. Materials such as plastics and metals will be discussed in a later section which deals with engineered barrier materials.

Carbon-14 and tritium can be mobilized via the processes of cellulose degradation, methanogenesis, bicarbonate utilization, denitrification sulphate reduction and degradation of aromatics. The more important processes will be discussed in the forthcoming sections.

6.3.1 Cellulose Degradation

Paper and cardboard are cellulosic in composition, being composed of long chain hexose units. Microbial degradation of cellulose can result in partial degradation of the six carbon members or splitting of a hexose unit, or complete degradation to carbon dioxide and water. Under aerobic conditions, there is a wide range of fungi and bacteria that can degrade cellulose; whereas, organisms capable of anaerobic breakdown are fewer in number, thus decreasing the potential for rapid degradation.

Energy requirements are higher for anaerobic degradation of cellulose. Nevertheless, once cellulosic degradation has been initiated during temporary storage, for instance, the degradation process will continue slowly over the lifetime of the repository. Cellulose will have no direct effect on radionuclide mobility, as the cellulose component of the waste is not radioactive. Nevertheless, the intermediate substrates (polysaccharides), carbon dioxide and water formed by total degradation of the cellulose will provide energy and nutrient sources for other microbial processes such as methanogenesis. Also, cellulose degradation may make C14 available which can be incorporated in carbon dioxide or water.

6.3.2 Methanogenesis

Methanogenesis is the microbial process by which carbon dioxide and water formed by aerobic degradation of cellulose recombine under anaerobic conditions to form methane. This process only occurs under anaerobic conditions and requires suitable nutrient and energy sources (CO_2, fatty acids and H_2). The production of methane depends upon the availability of water, temperature and pH states. An increase in

the moisture content results in increased production of methane gas[47]. Also, an increase in temperature to an optimum level of 35 - 42 C results in increased methanogenesis[48]. With respect to pH conditions within the repository, methanogenesis has only been observed within the pH range of 5.4 to 8.4. Hence, concrete engineered barriers would severely limit methanogenesis (if flooding occurs) due to the resulting alkalinity. This process can result in the formation of isotopes of water, carbon dioxide and methane, i.e., $^{3}H_2O$, $^{14}CO_2$ and $^{14}CH_4$, as well as other tritiated and ^{14}C hydrocarbons[49].

6.3.3 Bicarbonate Utilization

Apart from cellulosic degradation and methanogenesis, another important microbial process that can occur within the repository is bicarbonate utilization. As bicarbonate accounts for a large proportion of the inorganic carbon and ^{14}C inventory, the processes associated with its mobilization are important. This form of carbon can only be utilized by autotrophs[4]. Autotrophs are organisms that use carbon dioxide as the sole source of carbon for growth while energy for growth may be supplied by light or inorganic substances. One likely by product of bicarbonate utilization is methane gas. Also, the inorganic carbon source can be converted into an organic form. Nitrates are also capable of initiating the bicarbonate mobilization process utilizing Thiobacillus denitrificons. Nitrates may arise from the disposal of a waste form as cemented or bituminized salts. These nitrates would be subsequently leached from the waste form and be present in the aqueous phase as a source for microbes.

6.3.4 Organics

As commented in Section 6.2, organics can enter the repository via various direct or inadvertent means. Radionuclides existing in the waste form may be in either soluble or insoluble chemical forms. Complexing with various organics can greatly affect the mobilization of

the radionuclides. Many of the naturally occurring organic compounds are capable of forming stable complexes with radionuclides, and capable of supporting growth of aerobic and anaerobic micro-organisms. Apart from the host media and backfill materials, the waste forms also contain inorganic and organic compounds. Depending upon the environmental conditions, one may encounter autotrophs as well as heterotrophic microbial activities. Heterostrophs are organisms which require a source of exogenous organic compounds for growth and reproduction. Stimulation of heterotrophic microbial activities may affect the transformation and transportation of radionuclides. Reactions include oxidation - reduction, production of organic acids, synthesis of specific and non-specific sequestering agents, biodegradation of radionuclide organic complexes, bio-accumulation and remineralization of radionuclides, methanogenesis and production of radioactive gases.

Several classes of organic compounds consisting of organic acids, alcohols, aldehydes, amines, aromatic hydrocarbons, ethers and phenols have been identified in low-level waste disposal sites. Also present are synthetic chelating agents. In addition to synthetic chelating agents, complexing agents are found in decomposing organic matter introduced as part of the waste form and water mobile organic constituents, primarily humic and fulvic acid, present in the soil. Francis[39] has summarized a list of chemicals as shown in Table 12 that can result from the interaction of organics with micro-organisms. The significance of chelating agents in the mobilization of toxic metals and radionuclides is not clearly understood. Nevertheless, the presence of these compounds is of concern because of the potential for increased radionuclide mobility.

6.4 Microbial Interaction with Various Engineered Barrier Materials

6.4.1 Plastics

Plastics are a potential external engineered barrier material as well as a component of the waste form and repository drainage system

(geotextile). Plastics are normally considered to be highly recalcitrant materials and there is presently little information on their degradation under either aerobic or anaerobic conditons. Flooding or leaching of plastics removes the plasticizers, thereby increasing exposure of the polymer backbone to enzymatic and chemical action[37]. Nevertheless, it is anticipated that microbial decomposition of plastic is unlikely. Observations made at sanitary fill sites indicate that plastics remain impermeable to water after years of exposure to landfill leachate[37]. Regarding long term requirements for low-level waste containment, little information is available. One salient fact is that plastics will not hinder microbial activity.

6.4.2 Bitumen

Bitumen is presently being considered as an internal barrier/ buffer material and as a coating for external barrier materials. It is composed of a complex mixture of hydrocarbon compounds, some of which are water soluble and therefore could be leached. These compounds are biodegradable and will support microbial action. However, all the important metabolic pathways utilizing these compounds require aerobic conditions and, therefore, it is unlikely that these processes will continue after the repository is closed.

Very little information is available on the interaction of microorganisms with bituminized low-level waste. Most of the present research is on-going at Riso and Cadarache research laboratories. Microbiological experiments undertaken by Nomine' et al[50] at Cadarache indicate that the addition of bitumen increases the growth of certain families including nitrogen-fixing bacteria, fungi and actinomycetes, all of which are known for their ability to degrade complex hydrocarbons. This increase was noted under both aerobic and anaerobic conditions. The researchers also noted there was little

difference between the biological activity on the soil surface and that directly in contact with the waste form. Hence, it may be argued that the presence of bitumen does not appear to encourage the development of appreciable biological activity.

The studies undertaken by Brodersen et al[51] at Riso suggest that bitumen addition has no effect on the leaching rate of Cesium or Cobalt. The most significant impact of microbial degradation of bitumen may be the fact that the organic content of the pore fluid is increased by up to 20 fold. Also, the generation of carbon dioxide may have significant implications on the integrity of the disposal system. A recent study undertaken by Buckley et al[52] at CRNL appears to support the findings of the Riso and Cadarache laboratories in that the presence of bitumen does not significantly encourage microbial activity.

6.4.3 Epoxies/Resins

Pankhurst et al[53] have undertaken the most comprehensive study on the microbial attack of epoxy resins and polyurethanes. These experiments were undertaken under aerobic conditions only. Findings for individual organisms such as pseudomonas, serratia, streptomyces and dispergillus ranged from pronounced inhibition of growth to little growth. Epoxy resin hardener can be utilized as a food source by microbes. Overall, it was concluded that epoxy resins do not support appreciable microbial growth and therefore would be a suitable buffer material. These conclusions appear to be supported by on-going research programs being undertaken at Cadarache.

6.4.4 Concretes

Potential effects of microbial activity are widely reported in the literature for aerobic conditions. However, there is almost no information on concrete biodegradation under anaerobic conditions.

A general review of concrete hydration reactions has been presented in Section 4.2. Biodeterioration of concrete can be catalogued into 2 processes as follows:

a) Mechanical Processes - A common type of damage is repeated freezing and thawing of water present within the pores of the cement. Organisms growing on the surface of concrete indirectly increase the magnitude of each freeze-thaw cycle through their high water binding capacity.

b) Chemical Processes - Damage arises through the micro-organisms either mobilizing specific components of the material for nutrients, or energy (assimilatory damage), or by the secretion of corrosive or complexing metabolic products which chemically decompose the structure of concrete (dissimilating damage). The corrosion of concrete by sulphur oxidizing bacteria is a well known example of this phenomenon.

Many authors including Meyer et al[54] and Sand et al[55] have commented that sulphur-oxidizing bacteria (genus thiobacillus) dominate the corrosion of metals, limestone and concretes. This corrosion mechanism may occur due to backfill and geological host media interaction with the external surface of a concrete barrier, or an internal clay buffer system interacting with the internal face of a concrete barrier. Many natural clays contain high concentrations of reduced sulphur compounds such as sulphides, thiosulphides and tetrathionate. Two major groups of micro-organisms oxidize these compounds. An example of the first group is Beggiatoa alba. Hydrogen sulphide is oxidized in two steps to sulphuric acid:

$$2H_2S + O_2 \longrightarrow 2S + 2H_2O + \text{energy}$$
$$2S + 3O_2 + H_2O \longrightarrow 2H_2SO_4 + \text{energy}$$

The second group are the rod shaped bacteria of the genus Thiobacillus. Zajic[56] has described the metabolic behaviour of four important species, the first three present in an aerobic environment and the last, anaerobic. The four species are Thiobacillus thioparus, T. thioxidans, T. novellus and T. denitrificons.

The sulphuric acid attacks the physical structure of the concrete by reacting with calcium hydroxide to produce calcium sulphate. The end result is a disruption of the concrete matrix in the surface layer. Harboe[57] and Duddridge[58] has further noted that sulphuric acid reacts with aluminates to produce calcium sulphoaluminate (ettringite). This hydration product occupies a larger volume than the original aluminate and strains the matrix, causing cracking.

It should be noted that T. thioxidans and T. thioparus cannot grow in conditions of strong alkalinity and therefore fresh concrete is not colonized. If conditions occur to reduce the pH of the concrete surfaces, then it is possible for it to establish T. thioparus, which results in acidic metabolite excretion onto concrete. Subsequently, acid resistant species such as T. thioxidans can be established.

6.4.5 Metals

Under anaerobic conditions, sulphate-reducing bacteria like Desulfovibrio sp. and Desulfotomaculum sp. corrode iron. Postgate[59] has identified three main characteristics for this corrosion pathway:

a) restricted to anaerobic environments such as clay or waterlogged soils;

b) corroded metal is pitted rather than evenly corroded. Failure of iron structures caused by microbial activity tends to be through localized perforations rather than widespread deterioration;

c) the metal at the point of corrosion is graphitized.

In the presence of moisture, clean iron surfaces react with water and quickly become covered with a film of hydrogen. This depolarizes the surface and inhibits further reaction. Sulphate-reducing bacteria remove this layer by secreting an enzyme, hydrogenase, producing iron hydroxides and iron sulphide corrosion products. The proportion of hydroxide to sulphide is influenced by the concentration of organic compounds within the environment. Sulphide predominates in situations where organic material is high since hydroxide reacts with free hydrogen sulphide molecules. Postgate[59] also noted that areas of the metal surface subjected to alternating aerobic and anaerobic conditions corroded faster than in entirely anaerobic environments. To assess whether a soil is aggressive towards iron, redox potential and soil resistivity are better indicators than bacteria counts.

6.4.6 Backfill Materials

Mayfield and Barker[60] established the presence of microbiological activity in several backfill materials including Pembina Mountain clay, Avonlea oil well bentonite, Inco tailings, Domtar Sea Bond, Sodium Bentonite, Manitoba clay and Dresser Mineral bentonite. A wide range of microbial populations were observed for the different backfills and between different nutrient media. Bacteria species dominated the populations identified. Most were rod-shaped and varied from 1.0 um to 5.0 um in length. Fungi and yeasts were not numerous although

significant levels of actinomycetes were present. In most of the backfills, 80 percent of the bacteria were attached to soil particles, with the remaining 20 percent existing free within the material. The bacteria were not only spore forming types barely surviving under extreme conditions, but many were viable vegetative cells. Facultative anaerobics such as Bacillus sp. were also present.

TABLE 11: SUMMARY OF ENVIRONMENTAL FACTORS AFFECTING MICROBIAL GROWTH

FACTOR	GENERAL COMMENTS
1. NUTRIENTS	Nutrient sources required are carbon, nitrogen, phosphorus and sulphur sources, plus mineral and energy source. Heterotropic bacteria obtain structural and energy needs from organic carbon. Autotrophic bacteria require inorganic carbon (carbonates, dissolved CO_2) for growth and energy from light (phototrophic) or inorganics (chemotrophic).
2. WATER	Water is a prerequisite for microbial growth. Hygroscopic levels are sufficient. Presence of significant ionic content can reduce availability of water.
3. TEMPERATURE	Active microbial growth can extend from -15 to +90°C. Optimum temperature range is between 25 and 40°C.
4. OXYGEN AND Eh	Micro-organisms can exhibit aerobic, anaerobic or facultative anerobic types of metabolism, and hence can survive under a wide range of redox conditions. Anaerobic conditions considered to exist when oxygen concentration below 3×10^{-6} mol/L.
5. pH	Most micro-organisms do not tolerate extreme pH conditions. Optimum pH conditions for bacteria are 6 to 8 with fungi preferring an acidic environment.
6. RADIATION	Dosage required to kill 90 percent of population of vegetative bacterial cells is 300 to 400 Gy; whereas bacteria spores require a dose of 3 kGy to 4 kGy. Under anaerobic conditions, sensitivity of bacterial cells to radiation is reduced.

TABLE 12: ORGANIC COMPOUNDS IDENTIFIED IN DEEP AQUIFERS (Francis[39])

Phonols
Phenol
1,1 dimethylethy - 4 - methoxyphenol
(1,1 biphenyl) - 2 - ol
Organic Acids
2 ethylhexanoic acid
Heptanoic acid
Decanoic acid
Octadecanoic acid
Palmitic acic
Tetradecanoic acid
Several unidentified fatty acids
Low-molecular-weight fulvic acid
Others
Hydrocarbons, aldehydes and alcohols

7. HOST MEDIA - AN OVERVIEW OF GEOLOGICAL CONDITIONS IN SOUTHERN ONTARIO

7.1 Introduction

As outlined in Section 3, a variety of engineered barrier concepts have been investigated or proposed by AECL, Ontario Hydro, or others for the disposal of low-level wastes within near-surface Paleozoic and Quaternary sediments in southern Ontario. Concepts or options have included use of limestone caverns, where attenuating capacities for leachates may be quite variable; shallow land burial (SLB) in fine sands of moderately high hydraulic conductivity located above the local water table; and burial within fine grained shales or Quaternary clays and tills which have a comparatively greater potential for leachate retention, lower migration rates and chemical attenuation or renovation of escaped fluids containing contaminants.

In general, fine grained geologic materials provide a better back-up system for waste disposal and environmental protection for the following reasons:

a) lower travel time for escaped leachates;
b) general lack of producing water wells which may be affected;
c) large contact areas for physico-chemical processes due to high clay content, greater pore space etc.; and,
d) longer contact time between the contaminant and the geologic materials.

This section describes the sedimentary geological conditions in southern Ontario and assumes that with time, some contaminants will reach and interact with the host media.

7.2 Surficial Geology - Southern Ontario

7.2.1 General

Surficial materials within southern Ontario are of late Pleistocene and Holocene age, ranging from more than 125,000 years old to present day. The eldest of these are generally discontinuous, contain considerable stratified drift and commonly occur adjacent to Lakes Erie and Ontario. Because of the above points, these materials are generally not considered suitable for low-level waste disposal. Holocene materials deposited during the last 10,000 years consist of surficial peat deposits, shallow lacustrine sediments, thin - thick alluvium along streams and rivers, and postglacial eolian sands. Their high permeability are not suitable due to the eolian sands are under consideration in current waste disposal plans at CRNL because of non-availability of low permeability material. Consequently their disposal approach reflects different surficial geology.

In southern Ontario, potential low-level waste disposal sites include fine-grained Pleistocene sediments, which were deposited mainly between 15,000 and 12,000 years ago. These include fine- grained tills derived from lacustrine sediments and/or shale, fine-grained glaciolacustrine sediments, and combinations of the two. In eastern Ontario, fine-grained sediments are dominantly marine clays deposited between 12,000 and 10,000 years ago.

7.2.2 Tills

Fine-grained, near-surface till deposits most commonly occur adjacent to the Great Lakes Basins, where glaciers incorporated fine-grained lacustrine or glaciolacustrine sediments, or shales, and redeposited them as till. The following list shows the fine-grained tills which may be considered as candidates for waste disposal sites.

Till	Lake Basin	Redeposited Materials
Halton	Ontario-Erie	Shale, glaciolacustrine silt-clay
Port Stanley	Erie	Glaciolacustrine silt-clay (southern area only)
St. Joseph	Huron	Glaciolacustrine silt-clay
Rannoch	Huron	Glaciolacustrine silt-clay
Tavistock*	Huron-Georgian Bay	Glaciolacustrine silt-clay

7.2.3 Glaciolacustrine Sediments

Fine-grained glaciolacustrine silts and clay mantle a considerable area of southwestern Ontario. Of particular note are the extensive deposits of Glacial Lakes Warren and Whittlesey in the Brantford area and in the Sarnia-Windsor area. In some instances, these sediments are intercalated with fine-grained tills of suitable texture, which serve to increase the total thickness of the potential confining layer. These materials may be very thick, providing potentially good host media.

* Northern facies including overlying Mornington Till and associated sediments, and underlying Stirton Till and intervening sediments.

7.2.3.1 Clay Composition

Southern Ontario clays are comprised primarily of glacial rock flour with very few clay minerals representing diagenesis or post-depositional alteration. Though data on clay mineralogy and chemistry are scattered throughout many geological references, the most significant data sources are the reports published by G.R. Guillet[61]. Tables 13 and 14 provide mineralogic and chemical data abstracted from Guillet's reports[61,62]. The data are generally for late glacial and postglacial lacustrine sediments, some of which may be partially weathered and leached. As these sediments consist of rock flour, the ranges in mineralogical and chemical composition given probably encompass most glacial and glaciolacustrine clayey sediments in southern Ontario.

The chemical data indicate that more than 50 percent of the claysize materials consist of SiO_2, with the remainder distributed among several metallic and non-metallic compounds. The mineralogical composition indicates that non-clay minerals comprise slightly more than one-half of the "clay materials" with quartz being the dominant non-clay mineral. Clay minerals are dominated by illite, which occurs in moderate to abundant quantities; this is followed by chlorite, which occurs in minor amounts in most samples, though it ranges from not detectable to moderately abundant in occurrence. Expanding clay minerals occur in very minor quantities; these consist primarily of montmorillonite and interlayered minerals, with vermiculite occurring in small quantities. Swelling clays, occurring in southern Ontario, are frequently interpreted as the result of oxidation of chlorite, e.g., Quigley and Ogunbojeda[63], though some of these are undoubtedly derived from underlying shales.

7.2.4 Sands

Sand bodies are currently utilized at CRNL for waste disposal. Sands occur over large areas and in considerable thickness in two primary settings in southern Ontario:

a) As large deposits of lacustrine fine sands and silts deposited as end or interlobate moraines (e.g., Oak Ridge and Orangeville Moraines); and,
b) As large deltaic sand plains.

The former are usually high, dry, groundwater-recharge areas, with the water table at considerable depth, whereas, the deltaic sand plains frequently have their water table within 5 m of the ground surface.

7.2.5 Groundwater Levels and Hydraulic Conductivity

7.2.5.1 Clay and Clay Till

Hydraulic conductivities of fine-grained tills in southwestern Ontario were provided by Desaulniers et al[64] for four sites. These sites included Halton Till, St. Joseph Till, glaciolacustrine clay and perhaps the Rannoch Till. From field and laboratory observations, hydraulic conductivities were in the order of 10^{-10} to 10^{-9} m/s. Porosities ranged from 0.20 to 0.60, but were generally in the 0.30 - 0.40 range. Using these data, calculated average linear groundwater velocities ranged from approximately 13 - 26 m per 10,000 years. For fractured, desiccation crusts, hydraulic conductivities are increased by one or two orders of magnitude; these crusts are generally 2 - 3 m thick, but range up to 6 m in thickness.

Groundwater tables in clays and clayey tills are generally within 5 m of the surface. Thus, low-level waste sites developed in these materials would probably extend below the local water table.

7.2.5.2 Sands

Hydraulic conductivities of sands are highly variable; for example, materials in the Oak Ridge Moraine may be expected to vary from 4 x 10^{-6} m/s for silt to 5.7 x 10^{-5} m/s for fine sand, to 5 x 10^{-4} m/s for clean medium sand to 5.7 x 10^{-2} m/s for coarse sand and gravel.

7.3 Surficial Geology - Eastern Ontario

7.3.1 General

Low-level waste disposal in surficial materials in eastern Ontario includes disposal in sands, as is envisaged at CRNL, or perhaps disposal in Champlain Sea Marine clays which are comprised of relatively fine-grained soils.

7.3.2 Petawawa Sand Plain

The Petawawa sand plain consists of a series of fluvio-deltaic terraces, with an overlay of eolian fine sand. The geologic sequence is more complex than was originally believed[65]; the main sand body was emplaced about 10,500 years ago and the major terracing episodes occurred from that time to about 5,000 years ago. Eolian reworking of sands occurred following lowering of water levels from each terrace and prior to forestation; there are still active dunes in the area.

Burial of liquid radioactive wastes in shallow sand pits was initiated at CRNL in the mid 1950's. The disposal area is part of a recharge area of a shallow groundwater flow system containing two aquifers[66]. The deposit consists of several sand units with intervening and basal silt-clay layers. The sand mineralogy[66,67] is dominated by plagioclase feldspar and quartz (70 - 80%), the remainder consisting of minor amounts of numerous other minerals including sericite, mica, chlorite, K-feldspar and others. The silt and clay fractions of the middle silt-clay unit contains about 35% mica-vermiculite, 25% chlorite, 15% hornblende, 15% quartz and 10% plagioclase. Fines within the sand units are predominantly quartz and plagioclase with minor amounts of amphibole, K-feldspar, mica and interstratified mica-vermiculite[67]. Little geochemical weathering has occurred.

Hydraulic Conductivity: The sands have a mean porosity of 0.38, specific gravity of 2.73 gm/cm^3 and an estimated bulk density of 1.7 gm/cm^3. Hydraulic conductivities are in the order of 1×10^{-5} to 5×10^{-5} m/s[67]. During the past 25 years, radioactive plumes have developed within both aquifers. These have been separated using gas-liquid chromatography (GLC) into ^{90}Sr and ^{137}Cs plumes moving at a much lower velocity than the groundwater, i.e., the groundwater velocity is approximately 10^{-6} m/s; ^{90}Sr is about 3% that of groundwater and ^{137}Cs about 0.3% of that of the groundwater. Details of this experimental case are given in Jackson and Inch[67].

7.3.3 Champlain Sea Clay

Champlain sea clays may constitute potential host media for low- level waste. These are located within the Ottawa - St. Lawrence Valley and were deposited between 10,000 and 12,000 years ago. The marine clays

are up to 60 m thick, but thicknesses of less than 30 m are more common; a weathered crust up to 2 m thick is present. Clay size material is generally in the order of 70 - 90%. Average clay mineralogy and chemical composition on a regional scale are given in Tables 13 and 14.

Amorphous materials generally comprise an average of about 2 - 12% of the soil solids. Within the weathered surface crust, additional smectite may be present at the expense of the chlorite and biotite[69].

Groundwater Levels and Hydraulic Conductivity: The Champlain Sea clays have hydraulic conductivities in the order of 10^{-8} to 10^{-10} m/s and mean porosity of about 0.6. Water tables are very close to the surface and in the fissured surface crust, hydraulic conductivities are much greater than at depth.

7.4 Bedrock Geology

7.4.1 General

The Paleozoic (bedrock) geology of southern Ontario is divided into two areas (Figure 1) by Precambrian rocks of the Frontenac Arch, i.e., those rocks located south and east of Ottawa, and those rocks located south and west of Kingston. The former area is the Ottawa Embayment, which is part of the central St. Lawrence Lowland, while the latter comprises the western St. Lawrence Lowland containing the Michigan Basin on the northwest side of the Algonquin Arch and the foreland Allegheny Basin on the south. The strata are for the most part undeformed, but are locally

folded and faulted. In the Ottawa Embayment, normal faulting is widespread and the Paleozoic rocks are frequently in fault contact with Precambrian rocks[69].

7.4.2 Paleozoic Carbonate Rocks

Paleozoic carbonate rocks underlie much of eastern and southwestern Ontario as illustrated in Figure 1. These rocks have been suggested as possible sites for waste entombment in subsurface caverns. The geological suitability of this proposal depends on the level of back-up containment the rock medium is expected to provide.

In southern Ontario, groundwater flow within bedrock units is generally toward the southwest with some divergence toward either the Michigan on Appalachian structural basins. Hydraulic conductivities for the principal carbonate rock units and listed below. These are representative numbers only as well bedded, jointed, near surface rocks may have considerably higher conductivity.

Rock Types	Hydraulic Conductivities
Devonion Carbonates	10^{-5} m/s average
Silurian Carbonates	10^{-3} to 10^{-9} m/s (average 10^{-5} m/s)
Ordovician Carbonates	10^{-4} to 10^{-6} m/s

Carbonates in eastern Ontario may be expected to have similar ranges of hydraulic condutivity. However, as reported by Scott[71], thick massive sections within the Ottawa Formation (Group) may be relatively impermeable, possibly affording better containment opportunities than those sections with greater hydraulic conductivities.

The Paleozoic carbonate units are frequently utilized as aquifers for domestic water supplies. The selection of these rocks for waste emtombment may be expected to arouse greater environmental and social issues than rocks of lesser human interest, e.g. shales.

7.4.3 Paleozoic Shales

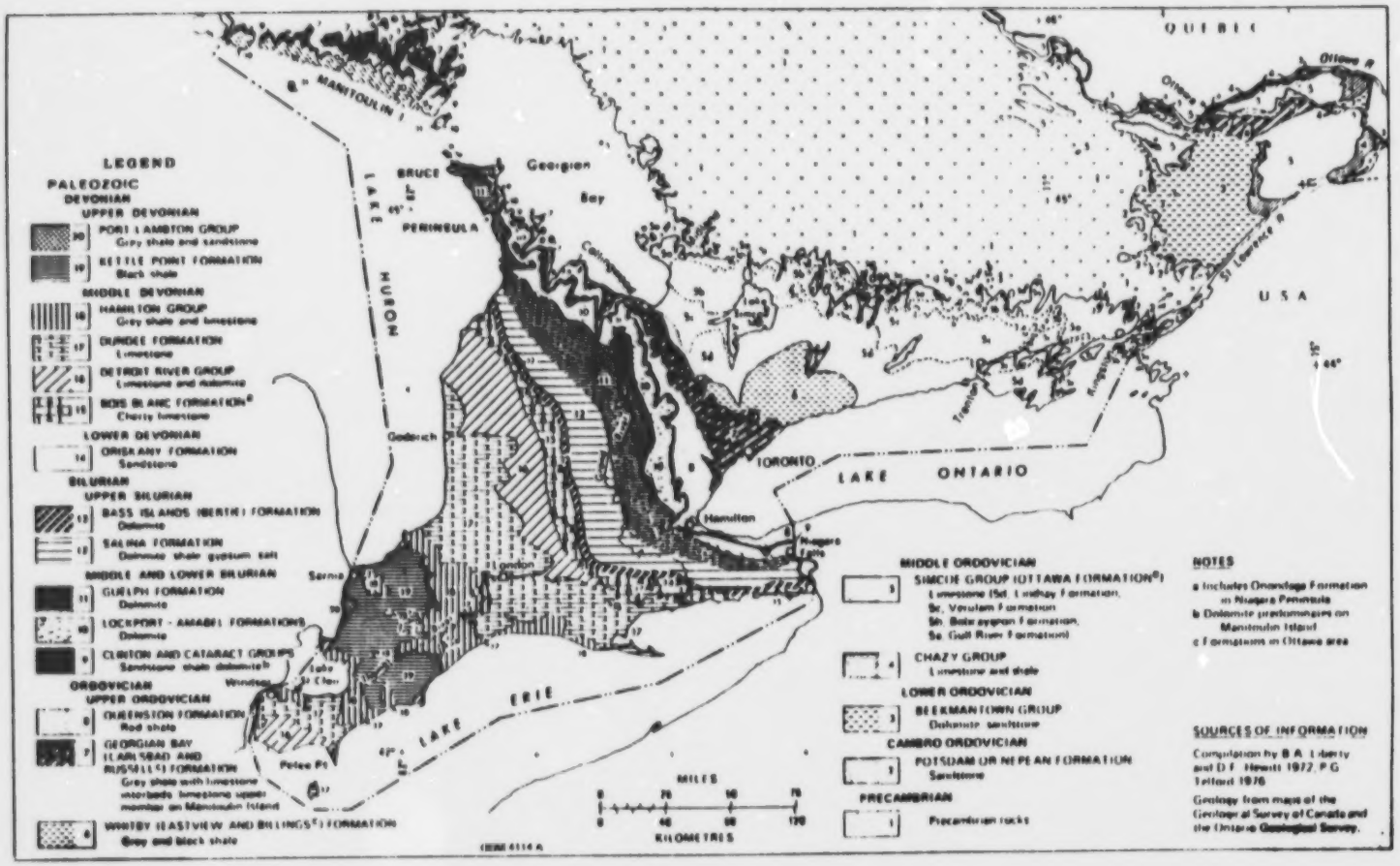
Paleozoic shales in southwestern Ontario generally provide a more suitable medium for low-level waste disposal if the rocks are expected to provide containment. Estimates of hydraulic conductivities for the shales are given below:

Rock Types	Hydraulic Conductivities
Devonion Shales	10^{-10} m/s average
Silurian Shales	10^{-9} to 10^{-10} m/s
Ordovician Shales	10^{-5} to 10^{-10} m/s

Conductivities for the fractured upper few metres of shale may range up to 10^{-5} m/s.

Shale chemistry and mineralogy are provided in Tables 16 and 17. These data are derived primarily from the reports of G.R. Guillet[61,62]. The data indicate that the major shale units contain more than 60% clay minerals which are dominated by illite and chlorite. Expanding clay minerals, mainly vermiculite and interlayered clay minerals, occur in several of the shale units in concentrations ranging from zero to 10 percent. Kaolinite occurs in small percentages within some of the Devonian shale units.

On a site-specific basis, the Paleozoic shales may be expected to provide sites with thick deposits having low hydraulic conductivities and relatively uniform chemical composition. These may provide a suitable containment medium for low-level radioactive wastes.



BEDROCK GEOLOGY OF SOUTHERN ONTARIO

FIGURE 1

TABLE 13: AVERAGE MINERAL COMPOSITION OF SOME SOUTHERN ONTARIO CLAYS (from G.R. Guillet[62,63])

	NON-CLAY MINERALS						CLAY MINERALS*		
	QUARTZ	CALCITE	DOLOMITE	NA - Ca FELDSPAR	K FELDSPAR	AMPHIBOLES	ILLITE	CHLORITE	EXPANDING MINERALS
Occurance	84	84	84	84	84	84	84	84	84
Mean (%)	28	11	6.4	4.1	1.31	1.0	B+	C+	C-
Range (%)	18-64	0-44	0-32	0-15	0-5	0-5	C-A	ND-B	ND-A

* A = abundant; B = moderate; C = minor; ND = not detected

TABLE 14: AVERAGE CHEMICAL PROPERTIES FOR SOME SOUTHERN ONTARIO CLAYS (from G.R. Guillet[62,63])

	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	TiO_2	CO_2	H_2O+	H_2O-	SO_3	LOI
Occurance	21	21	21	21	21	21	21	20	20	20	20	21	21
Mean (%)	49.8	11.2	4.8	11.8	3.6	1.1	2.75	0.59	10.0	2.32	1.3	0.34	14.2
Range (%)	37 - 65	7.9 - 15.2	3.1 - 7.2	1.1 - 27.7	1.4 - 8.2	0.6 - 2.0	2.3 - 4.0	0.4 - 0.91	0.7 - 19.3	1.5 - 3.6	0.5 - 2.0	0.1 - 0.7	6.7 - 22.0

TABLE 15: AVERAGE MINERAL COMPOSITION OF SOME EASTERN ONTARIO CLAYS (from G.R. Guillet[62,63])

	NON-CLAY MINERALS						CLAY MINERALS - ABUNDANCE*			
	QUARTZ	CALCITE	DOLOMITE	NA - Ca FELDSPAR	K FELDSPAR	AMPHIBOLES	PERCENT	ILLITE	CHLORITE	EXPANDING** MINERALS
Number of Occurance	18	18	18	18	18	18		18	16	17
Mean (%)	18.7	1.6	2.1	6.7	1.1	5	65	A	B-C	C
Range (%)	12-29	0-6	0-7	2-9	0-3	1-10		A-C	A-D	A-D

* A = abundant; B = moderate; C = minor; D = trace
** Interlayered and montmorillonite

TABLE 16: AVERAGE CHEMICAL COMPOSITION OF EASTERN ONTARIO CLAYS (from G.R. Guillet[62,63])

	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	Na_2O	K_2O	TiO_2	CO_2	H_2O+	H_2O-	SO_3	MnO	LOI	SOLUBLE SALTS
Number of Samples Tested	2	2	16	16	2	2	2	2	2	2	2	2	1	16	15
Mean (%)	54.9	16.2	7.2	3.7	4.2	2.6	3.4	0.8	1.0	2.5	2.5	0.4	0.1	7.0	1.3
Range (%)	53 - 56	16.1 - 16.4	5.2 - 8.2	2.2 - 8.9	3.7 - 4.6	2.5 - 2.6	3.1 - 3.6	0.7 - 0.9	0.1 - 2.0	2.2 - 2.8	1.8 - 3.2	0.3 - 0.5	--- -	4.6 - 9.2	0.5 - 2.6

TABLE 17: MINERALOGY OF PALEOZOIC SHALES IN SOUTHERN ONTARIO

FORMATION OR GROUP	NON-CLAY MINERALS					CLAY MINERALS - ABUNDANCE*				
	QUARTZ %	FELDSPAR %	CALCITE %	DOLOMITE %	OTHER %	TOTAL %	ILLITE	CHLORITE	EXPANDING	NO.
KETTLE POINT	32	1	1	ND	-	65	A	B	ND	1
HAMILTON	16-29	0-6	6-52	0-16	1	63	A-B	A-C	ND-C	10
SALINA	27	0-2	5-20	36-37	-	35	B-C	C	ND	2
ROCHESTER	12-16	0-1	13-39	19-29		40	A-B	C	C	3
CABOT HEAD	17-41	0-1	0-3	0-13	-	60	A	B-C	ND	12
QUEENSTON	12-34	0-9	2-30	0-20		60	A	B-C	ND-C	45+
GEORGIAN BAY	23-36	2-6	.5-29	0-5	-	60	A-B	B	ND-C	20+
WHITBY:										
Upper (Blue Mtn.)	19-28	0-3	1-2	0-4	-	70-75	A	A-B	ND-Tr	4
Middle (Gloucester)	26-27	0-2.5	ND	ND	-	70-72	A	B	Tr	2
Lower (Collingwood)	12-14	1-2	35-50	1-8	-	35-45	A-B	B-C	ND	2
SHADOW LAKE	6.0	4.5	6.0	59.0	-	24.5	B	C	-	1

* A = abundant; B = moderate; C = minor; Tr = trace; ND = not detected.

8. ANTICIPATED LONG TERM PERFORMANCE OF BARRIER MATERIALS FOR LOW-LEVEL WASTE

8.1 Construction and Post-Closure Conditions

Several distinct environments are likely to exist within a low-level radioactive waste repository during the expected lifespan of up to 500 years. During construction and subsequent emplacement stages, the repository will be open to the atmosphere and hence conditions will be aerobic. During this period, it is possible that organic matter, micro-organisms and water (rainfall) may be directly or indirectly introduced into the waste repository. Colonization of micro-organisms will commence, with subsequent degradation of waste forms.

Once closure of the waste repository has been completed, aerobic conditions will continue until the available oxygen has been depleted. At the present time, no information exists on the length of time required for depletion of oxygen to occur. Nevertheless, during this period, aerobic micro-organisms will be displaced by anaerobic and facultative anaerobic micro-organisms. Processes such as methanogenesis and acid formation will be initiated. The rate of growth will not only depend upon the availability of nutrients, but also the temperature within the repository. High rates of anaerobic activity could result in an increase in internal temperature, thereby accelerating the rate at which engineered barrier materials would degrade.

Over the next several decades, the integrity of engineered barriers may decline. Humidity in the repository may increase with groundwater intrusion occurring. Groundwater intrusion will recontaminate the waste form and provide nutrients for microbial growth.

Gases such as carbon dioxide and methane are likely to be formed. Also, the degradation of organic materials may result in the formation of a variety of organic compounds, including chelating agents which will enhance the mobility of radionuclides.

8.2 Engineered Barrier Materials

8.2.1 General

For low-level waste disposal, barriers should remain effective for at least the period of institutional control, which is an undefined period of between 100 and 500 years. This time span is well beyond the experience history for engineered barriers presently in use for waste containment. Hence, predictions about long-term performance of the various materials under consideration must be approached with caution. This section addresses the performance anticipated for materials proposed by the major low- level waste producers, as well as other materials which have been utilized in industrial or municipal waste containment, but which may not have been specifically identified by the producers as candidate barrier materials.

Table 18 lists the various engineered barrier materials that have been discussed in Section 4 and 5 of the report. The anticipated long term performance of these materials under the assumed operating environment is indicated in the table.

8.2.2 Natural Soils and Soil Mixes

8.2.2.1 Clayey Soils

Natural clayey soils used as backfill and in external barriers and covers should have as high a clay mineral content as possible to provide

desirable properties of low hydraulic conductivity and high cation exchange capacity. Suitable clay minerals, in general order of increasing permeability, are sodium montmorillonite, calcium montmorillonite, illite and kaolinite.

Although a clay barrier can be constructed at an initial hydraulic conductivity of 10^{-10} m/s, or less, physical and chemical processes can lower the effectiveness of the barrier with time. Detrimental processes include:

a) formation of cracks due to seasonal drying and freeze-thaw processes;
b) penetration of the barrier by roots and burrowing rodents;
c) differential settlement causing cracking;
d) changes in pore water chemistry and ionic composition which increase the porosity and permeability of the soil;
e) chemical dissolution of the clay minerals by acids and bases, leading to increased permeability; and,
f) gas generation.

The chemical constituents of low-level waste are generally thought to be sufficiently dilute not to present any likelihood of significant increase in the permeability of a clay barrier due to changes in soil and porewater chemistry, or ionic exchange. However, potential cracking and penetration of the barrier due to processes a), b) and c) could lead to a significant impairment of barrier effectiveness with time. These processes could be particularly detrimental to the performance of the cover, with the degree of effect decreasing with increase in the depth of burial.

8.2.2.2 Soil Mixes

Soil admixtures which may be considered for use in engineered barriers include bentonite-sand, sand-clay, soil asphalt and soil cement. All of these admixtures can be designed and constructed such that the initial permeability of the placed material is similar to that of natural clay soils. However, the same processes which can be detrimental to the performance of clay barriers will also affect soil admixtures.

a) Bentonite Mixes: Bentonite admixtures have the advantage of containing montmorillonite, which has both low permeability and a high cation exchange capacity, thus serving a dual purpose of reducing seepage and adsorbing chemicals leached from the waste repository. Sodium montmorillonite is preferred, as it has the best absorption capacity for both water and chemicals. Disadvantages of bentonite admixtures are difficulty with uniform blending of the soil (usually sand) and bentonite and ensuring a high quality integral barrier. Also, drying can cause shrinkage cracks to form.

b) Sand-Clay: Sand-clay mixtures will behave essentially as natural clays, except that they will tend to be less sensitive to cracking from drying, as the structure will tend to be denser and the clay content is lower than for a clay only barrier. The main disadvantage of a sand-clay mixture is the difficulty of achieving a uniform, consistent blend. Also, the capacity to absorb waste chemicals would be reduced in proportion to the reduced amount of clay used in the mix.

c) Soil Cement and Soil Asphalt: Documented instances of the use of soil cement and soil asphalt linings in industrial and municipal landfills are very few. This, together with concerns about brittleness and susceptibility to cracking would likely

render these materials unsuitable for use in external barriers, although there may be potential applications as buffers, or internal barriers.

8.2.3 Cement

This section addresses the factors which will affect the long term performance of concretes and cements, with and without admixtures. Physical properties, freeze-thaw resistance and microbial effects are discussed. Also, information is provided on the use of coatings.

8.2.3.1 Physical Properties

It is anticipated that the compressive strength of concrete barriers with and without admixtures will not be an issue. The permeability of the barriers is of more importance. The permeability of intact concretes and cements is in the region of 10^{-11} to 10^{-13} m/s. The addition of admixtures can reduce the permeability to 10^{-15} m/s. Gas permeability values are in the region of 2 to 3 orders of magnitude higher. Although these permeability values are lower than observed for clays, cracking can occur for several reasons including shrinkage, foundation settlement, frost effects and earthquake shocks. The addition of admixtures may or may not make the concretes more durable. For instance, research on cements with silica fume addition has shown that shrinkage depends on the relative humidity state. At low humidity values, major fracturing of the cements was observed.

8.2.3.2 Freeze-Thaw Resistance

An extensive study was undertaken by Virtanen[72] on the freeze-thaw resistance of concrete with and without admixtures of silica fume, blast furnace slag and fly ash. The following conclusions can be drawn:

a) The air content of the concrete has the greatest influence on freeze-thaw resistance.

b) The addition of blast furnace slag, silica fume and fly ash reduces the amount of entrained air.

c) Silica fume addition improves freeze-thaw resistance as compared to concrete with the same strength and air content; whereas, the addition of blast furnace slag or fly ash results in little or no increase in freeze-thaw resistance.

d) Replacement of a major part of the cement by these admixtures can have a detrimental effect on freeze-thaw resistance.

8.2.3.3 Microbial Effects

As discussed in Section 6, microbial degradation of the waste form through processes such as cellulose degradation, methanogenesis, bicarbonate utilization and organic degradation can result in the production of gases such as carbon dioxide and methane. In the presence of carbon dioxide, concretes and cements undergo carbonation, which entails transformation of the hydration product calcium hydroxide to calcium carbonate (calcite). The rate of carbonation depends upon the gas pressure and the relative humidity. Carbonation results in a change in the pore size distribution of the concrete and cement, with a larger portion of large radii pores. The total pore volume of the carbonated portion of the concrete is larger than for the non-carbonated, with a corresponding decrease in the compressive strength. Carbonation studies undertaken on oil well cements by one of the report authors indicate that the outer pores become plugged by the calcite, thus restricting the diffusion of CO_2 into the inner pores. This pore plugging results in a decreased "apparent" permeability. Blended cements have a finer pore

structure and therefore reduce the effect of carbonation. The pore plugging mechanism associated with carbonation appears to have a modulating effect on shrinkage. Long term effects of carbonation on strength and permeability are not known and cannot be assessed unless the amount of CO_2, pressure and relative humidity state within the repository is known.

The environment within a concrete repository will be highly alkaline, i.e., pH = 10 - 11. This alkalinity will tend to severely reduce microbial growth. Nevertheless, specific genera of micro-organisms will produce acids such as sulphuric and organic acid from the degradation of organics. Over a long period of time, production of these acids will reduce the pH of the repository environment, resulting in the formation of calcium sulphate from the hydration product calcium hydrate. The formation of calcium sulphate has the tendency to disrupt the concrete matrix. Also, Harboe[57] and Duddridge[58] have noted that sulphuric acid reacts with aluminates to form ettringite. This mineral occupies a large volume, resulting in cracking of the concrete.

Reinforcement would also be susceptible to corrosion from the acid environment. According to Gjorv[73], the addition of admixtures such as silica fume can make the concrete sulphate resistant. Otherwise, sulphate resistant, Type CSA 20, concrete can be used.

8.2.4 Bitumen

Bitumen has been considered both as a protective coating for concrete and as an internal buffer material. Bitumen can be degraded by various micro-organisms, resulting in the production of CO_2. The rate of degradation is extremely slow and dependent upon the pH. In a highly alkaline environment associated with concrete, bitumen degradation will be exceedingly low.

Apart from CO_2 production, microbial degradation of bitumen results in a high organic concentration in the pore water. These organics can significantly increase the mobility of the radionuclides.

As a protective coating, the bitumen would ensure a non-alkaline environment. This environment would enhance both microbial activity (degradation of waste forms) and bitumen degradation and hence, would be considered negative to the overall performance of the barrier. It is anticipated that the process of drying with corresponding shrinkage, would result in cracking of the bitumen layers and eventually forming a powder. Freeze-thaw cycling resulting from temperature changes within the repository will have the same detrimental results. If bitumen is to be used within the repository, concrete should be used as the engineered barrier to maintain the alkaline environment. The use of a biocide may also be considered, but would only be effective in the short term. External use of a coating barrier material may be beneficial in terms of restricted movement of fluid from the geomedia.

8.2.5 Polymeric Membranes

The use of polymeric membranes for containment of both municipal and industrial liquid wastes has increased dramatically in recent years, with generally favourable experience being gained with the more durable, chemically resistant compounds, such as high density polyethylene (HDPE), hypalon and neoprene. However, there are several concerns about the utilization of polymeric membranes in engineered barriers for low-level waste disposal. The main concerns are susceptibility to penetration by roots and burrowing animals, tearing due to differential settlement, chemical and microbial attack and loss of strength and polymer breakdown over time.

There is no assurance that the effective life span of polymeric membranes would be much more than about 50 years. In fact, Mezga[74] suggests a useful lifetime of only 25 years. For these reasons, it would be imprudent to rely on polymeric membranes as a primary external barrier, although there may be a role as a backup liner within a multi-barrier system.

8.2.6 Drainage and Protective Materials

Layers of granular soil, possibly combined with geotextiles may form part of the barrier system, particularly the cover. Comments on potential materials are provided below.

8.2.6.1 Coarse-Grained Soils

Natural coarse-grained soil consisting of gravel, cobbles and boulders may be placed as part of the cover system to protect the underlying low permeability barrier. Protection would be provided against erosion, root penetration, burrowing animals and traffic loads. If the layer is sufficiently thick, it would also offer some protection against freezing and drying effects in the underlying low permeability cover unit. Coarse- grained materials, if consisting of hard, durable rock particles, could have considerable longevity and resistance to breakdown within the life of the repository.

8.2.6.2 Geotextiles

Geotextile fabrics have been proposed for incorporation into the cover sytem, where they would serve a combined function of a drainge and separator layer between different soil types, and as reinforcement of the cover to withstand differtial settlement effects. While the short

term benefits of fabrics are evident, there is little or no evidence available that the materials would not degrade and lose much of their strength with time. They could be particularly susceptible to chemical breakdown and breaching by animal and vegetative intrusion. The main advantage of using geotextiles would be to retard the eventual deterioration of the repository.

8.3 Geological Host Media

An overview of near surface geological conditions in southern Ontario, where low-level waste repositories are under consideration, was presented in Section 7. The geologial host materials will function either as a primary containment medium, or as a back-up, depending on the overall design of the repository system. Geological materials currently under consideration for disposal sites include clay and clay till, sand, shale and limestone.

The general suitability of each material type being considered is discussed below. In general, fine grained geologic materials provide a better back-up system for waste storage and environmental protection for the following reasons:

a) lower travel time for escaped leachates;
b) general lack of producing water wells which may be affected;
c) large contact areas for physico-chemical processes due to high clay content, greater pore space, etc.; and,
d) longer contact time between the contaminant and the geologic materials.

8.3.1 Clay and Clay Till

Both clay and clay till deposits in southern Ontario have engineering properties which are generally conducive to the retention of waste materials. Below the zone of surface weathering and fissuring, permeabilities are low (10^{-9} m/s range) and if a substantial component of illite clay mineral is present, the capacity for absorbing waste ions and radionuclides is considerable.

The main disadvantage associated with clays as host media is that the groundwater table may be relatively close to the ground surface. Over the life span of the respository, it may be difficult to ensure that fluctuations in groundwater levels will not reach the repository at some time. Flooding of the low-level waste would greatly accelerate biological processes which could lead to deterioration of the primary barriers and an accelerated release of contaminants.

8.3.2 Sand

Sand may have a permeability of from about 5×10^{-6} m/s to as high as 5×10^{-2} m/s. Corresponding contaminant migration rates could therefore be several orders of magnitude higher than in clay or clay till. On the other hand, sand deposits within areas of topographic highs may have a relatively low water table and there may be a smaller likelihood that water could rise in the future to saturate the repository. Sands may exhibit some adsorption capacity if they possess amorphous coatings.

8.3.3 Bedrock

Both shale and limestone are under consideration for siting low- level waste repositories. Near surface shale is likely to exhibit properties

which would be similar to those of clay and clay till, except that fractures (joints and faults) would constitute potential zones of seepage which may be highly conductive. Water table fluctuations and the possibility of saturation of the repository over the long term are concerns that would have to be examined in the design of a barrier system in shale.

Near surface limestone is often fractured and can exhibit relatively high permeability, thus limiting its usefulness as a barrier medium. The tightest rock conditions will probably be experienced at depth (below 10 - 20 m), but below the water table. As domestic water wells frequently are located within limestone aquifers and in view of the likelihood of significant fracture permeability, limestone may not offer a particularly suitable host medium for disposal of low-level waste. In spite of this concern, relatively tight zones do occur in limestone and may offer potential disposal sites.

TABLE 18

ENGINEERED BARRIER MATERIALS - GENERAL PERFORMANCE EVALUATION

MATERIAL	Past applications in engineering barriers	Trafficability	Seepage resistance	Erosion resistance	Desiccation resistance	Freeze/thaw resistance	Crack resistance	Stability on steep side-slopes	Vegetation Support	of construction	Longevity (>100 years)	Resistance to biological deterioration	Resistance to rodent burrowing	Resistance to root penetration	Radon gas control	Absorption of gamma waves
Concrete	G	G	G	G	G	F	G	G	G	F	F	F	P	G	G	G
Clayey soil	G	P	G	F	P	F	P	G	P	G	F	G	G	P	G	G
Bentonite	G	P	F	F	F	F	P	G	P	G	F	G	P	P	F	G
Sand-bentonite admixtures	G	P	G	F	F	F	P	G	P	G	F	G	F	P	G	G
Gravel, cobbles and boulders	G	G	P	G	G	G	G	G	G	G	G	G	P	F	P	G
Soil cement	F	P	G	G	G	P	G	G	F	F	P	G	P	P	G	G
Soil asphalt	F	P	G	G	G	F	G	G	F	F	P	P	P	P	G	G
Spray-on asphalt emulsion	F	P	G	G	G	G	P	P	P	F	P	P	P	P	G	P
Hydraulic asphalt concrete (HAC)	F	G	G	G	G	G	G	G	G	F	P	P	P	P	G	P
Polymeric membranes: (CPE, hypalon, neoprene, CSPE, HDPE, PVC)	F	P	G	G	G	G	P	P	P	F	P	P	P	P	P	P
Geotextiles (drainage and filter layers)	F	G	P	G	F	G	G	G	F	G	P	P	F	P	P	P

G = good performance; F = fair to neutral performance; P = poor performance.

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